The structure of fluctuations near mean-field critical points and spinodals and its implication for physical processes

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We analyze the structure of fluctuations near critical points and spinodals in mean-field and near-mean-field systems. Unlike systems that are non-mean-field, for which a fluctuation can be represented by a single cluster in a properly chosen percolation model, a fluctuation in mean-field and near-mean-field systems consists of a large number of clusters, which we term fundamental clusters. The structure of the latter and the way that they form fluctuations has important physical consequences for phenomena as diverse as nucleation in supercooled liquids, spinodal decomposition and continuous ordering, and the statistical distribution of earthquakes. The effects due to the fundamental clusters implies that they are physical objects and not only mathematical constructs.

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I. INTRODUCTION

Systems that are mean-field or near-mean-field are common in nature. Examples of such systems include metals with long-range elastic forces [1, 2], earthquake faults with long-range stress transfer Green's functions [3], and polymers [4]. The connection between the range of the interaction and mean-field behavior was made by Kac and collaborators [5] who noted that a system with a pairwise additive potential of the form

$$V(x) = V_R(x) + \gamma^d \Phi(\gamma x), \tag{1}$$

becomes mean-field in the limit $\gamma \to 0$. In Eq. (1) $x = |\mathbf{x}|$, $V_R(x)$ is a short range reference potential, and d is the spatial dimension. The limit $\gamma \to 0$ is taken after the thermodynamic limit and before a critical point is approached. It is also required that [5]

$$\int d\mathbf{x}\, \gamma^d |\Phi(\gamma x)| < \infty. \tag{2}$$

so that the energy per particle or spin remains finite in the $\gamma \to 0$ limit. The interaction range R is defined by the second moment of the potential,

$$R^2 \propto \int d\mathbf{x}\, x^2 \gamma^d \Phi(\gamma x) \propto \gamma^{-2}. \tag{3}$$

Hence, as $\gamma \to 0$, $R \to \infty$. We will refer to systems with $R \gg 1$ but not infinite as *near-mean-field*; systems with $R \to \infty$ are *mean-field* [6].

The kinetics of phase transitions is different in systems with $R \gg 1$ than in systems with $R \sim 1$. For example, nucleation in the former often occurs near a pseudospinodal [7, 8, 9] where the surface tension is small, which results in a nucleating droplet that has a different structure [10, 11, 12, 13, 14, 15, 16, 17] than that near the coexistence curve in systems with $R \sim 1$ [18, 19].

In addition, the early stage growth of the peak of the equal time structure function during continuous ordering and spinodal decomposition in systems with $R \gg 1$ is described by the Cahn-Hilliard-Cook (CHC) theory [20, 21, 22] for a time proportional to $\ln R$ after the quench [4]. The morphology of the early stage evolution differs from that in systems with $R \sim 1$ [23, 24, 25] for which there is no time interval when the CHC theory is applicable [19].

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The mean-field limit of several earthquake fault models can be described by an equilibrium theory [3, 26, 27]. In near-mean-field systems the smaller earthquake events are related to fluctuations about the free energy minimum near the pseudospinodal [3].

In these and other examples the structure of the fluctuations near the mean-field critical point and the pseudospinodal is important for understanding the behavior of the system. In this paper we analyze the structure of the fluctuations and its relation to the underlying clusters. We use field theory, scaling arguments, and cluster analysis and relate the structure of the fluctuations to the nature of nucleation, the possible existence of a pseudospinodal in supercooled fluids, and the behavior of the models of earthquake faults. The results of simulations done to test the predictions are also discussed.

In Secs. II and III we discuss the Landau-Ginzburg theory [28] and the Parisi-Sourlas [29, 30] approach based on the Langevin equation with random Gaussian noise to study fluctuations near mean-field critical points. We use the same field theory techniques in Sec. IV to discuss fluctuations near the spinodal. In Sec. V we discuss the fluctuation morphology for mean-field and near-mean-field systems. In Sec. VI we use the Landau-Ginzburg and Parisi-Sourlas approaches to discuss the relation of the fluctuations to the clusters. We examine the relation between the fluctuation structure and spinodals in supercooled fluids in Sec. VII and discuss the relation between the fluctuation structure and nucleation in Sec. VIII. In Sec. IX we relate the cluster structure to cellular automata models of earthquakes. We summarize our results and discuss future work in Sec. X. The mapping of thermal systems onto percolation models is discussed in Appendix A.

Our main results include the following. (1) There exist objects, which we call fundamental clusters, that have a density and lifetime dependence that is very different from the scaling of the density and lifetime of the fluctuations. The fundamental clusters are defined by the mapping of the critical point (or spinodal) onto a percolation transition. This difference is in contrast to non-mean-field systems where the clusters are geometrical realizations of the fluctuations. (2) The fundamental clusters are physical objects that have measurable consequences, which are explored for earthquake fault models, nucleation, and the measurement of the (pseudo)spinodal in near-mean-field systems.

II. SCALING OF ORDER PARAMETER FLUCTUATIONS

We first discuss (near-)mean-field systems from the perspective of field theory based on the Landau-Ginzburg-Wilson Hamiltonian [31, 32]

$$H(\phi) = \int d\mathbf{x} \Big[\frac{R^2}{2} [\nabla \phi(\mathbf{x})]^2 + \epsilon \phi^2(\mathbf{x}) + \phi^4(\mathbf{x}) - h\phi(\mathbf{x}) \Big]. \tag{4}$$

Without loss of generality we have set the proportionality constant in Eq. (3) equal to one. The partition function Z is

$$Z = \int \delta\phi \, e^{-\beta H(\phi(\mathbf{x}))}, \tag{5}$$

and the probability of the order parameter density $\phi(\mathbf{x})$ is

$$P_B(\phi) = \frac{e^{-\beta H(\phi)}}{Z}. \tag{6}$$

where $\beta = (k_B T)^{-1}$, T is the absolute temperature, and k_B is Boltzmann's constant.

Because we are interested in (near-)mean-field systems, we scale all lengths with R. We first discuss the critical point and defer our discussion of the spinodal to Sec. IV. To study the critical point we set $h = 0$ and assume that

$$\epsilon = \frac{T - T_c}{T_c} \ll 1, \tag{7}$$

where T_c is the critical temperature. For $\epsilon \ll 1$ and $h = 0$ we can use scaling arguments. It is straightforward to see from Eq. (4) that

$$H(\tilde{\phi}) = R^d |\epsilon|^{2-d/2} \int d\mathbf{y} \Big[\frac{1}{2} \big[\tilde{\nabla} \tilde{\phi}(\mathbf{y}) \big]^2 \pm \tilde{\phi}^2(\mathbf{y}) + \tilde{\phi}^4(\mathbf{y}) \Big], \tag{8}$$

where $\mathbf{y} = \mathbf{x}/R\epsilon^{-1/2}$, $\tilde{\phi}(\mathbf{x}) = \epsilon^{-1/2}\phi(\mathbf{x})$, $\tilde{\nabla} = R\nabla$, and the $+$ $(-)$ sign corresponds to $\epsilon > 0$ $(\epsilon < 0)$. We take $\epsilon > 0$ in this section except where otherwise noted.

The integral in Eq. (5) can be evaluated using saddle point techniques for $R^d \epsilon^{2-d/2} \gg 1$. We can give this requirement a physical meaning from the Ginzburg criterion [28], which states that a system can be considered to be mean-field if the mean square fluctuations of the order parameter are small compared to the square of the order parameter [32]. The order parameter ϕ is given by $L^{-d} \int d\mathbf{x} \, \phi(\mathbf{x})$, where L is the linear dimension of the system and ϕ corresponds to the magnetization in the Ising model.

The correlation length ξ is proportional to the linear spatial extent of the order parameter fluctuations. The mean square fluctuations in the order parameter are characterized by $\xi^d \chi$,

where χ is the isothermal susceptibility [32]. Near a mean-field critical point we have [33]

$$\xi \sim R\epsilon^{-1/2} \tag{9a}$$

$$\phi \sim |\epsilon|^{1/2} \qquad (\epsilon < 0) \tag{9b}$$

$$\chi \sim \epsilon^{-1}. \tag{9c}$$

(Equation (9b) is derived following Eq. (56).) The Ginzburg criterion requires that

$$\frac{\xi^d \chi}{\xi^{2d}\phi^2} \to 0. \tag{10}$$

If we substitute the scaling forms in Eq. (9) into Eq. (10), we obtain [4]

$$G = R^d \epsilon^{2-d/2} \to \infty. \tag{11}$$

We will refer to $G = R^d\epsilon^{2-d/2}$ as the Ginzburg parameter. In the limit $G \to \infty$ the system is mean-field. The system is near-mean-field for $G \gg 1$ (but finite). The latter criterion implies the well known result that the upper critical dimension at the critical point above which the system has mean-field critical exponents for all R, including $R \sim 1$, is four [32].

From Eqs. (6) and (8) we have

$$P_B(\tilde{\phi}) = \frac{\exp\left\{-\beta R^d \epsilon^{2-d/2} \int d\mathbf{y} [\frac{1}{2}[\tilde{\nabla}\tilde{\phi}(\mathbf{y})]^2 + \tilde{\phi}^2(\mathbf{y}) + \tilde{\phi}^4(\mathbf{y})]\right\}}{Z}. \tag{12}$$

For $G = R^d\epsilon^{2-d/2} \gg 1$ the Hamiltonian in Eq. (4) can be approximated by a Gaussian [32]:

$$P_G(\tilde{\phi}) = \frac{\exp\left\{-\beta R^d \epsilon^{2-d/2} \int d\mathbf{y} \frac{1}{2}[\tilde{\nabla}\tilde{\phi}(\mathbf{y})]^2 + \tilde{\phi}^2(\mathbf{y})\right\}}{Z_G}, \tag{13}$$

where Z_G is the functional integral over $\tilde{\phi}$ of the numerator in Eq. (13). We use $P_G(\tilde{\phi})$ to calculate the structure function $S(\tilde{k})$:

$$S(\tilde{k}) = \epsilon <\tilde{\phi}(\tilde{\mathbf{k}})\tilde{\phi}(-\tilde{\mathbf{k}})> = \frac{\epsilon \int \delta\tilde{\phi}(\tilde{\mathbf{k}}) \exp[-\beta R^d \epsilon^{2-d/2} \int d\tilde{\mathbf{k}} (\tilde{k}^2 + 1)\tilde{\phi}(\tilde{\mathbf{k}})\tilde{\phi}(-\tilde{\mathbf{k}})]\tilde{\phi}(\tilde{\mathbf{k}})\tilde{\phi}(-\tilde{\mathbf{k}})}{Z_G}, \tag{14}$$

where $\tilde{\mathbf{k}} = R\epsilon^{-1/2}\mathbf{k}$ and $\tilde{\phi}(\mathbf{k}) = \tilde{\phi}(-\mathbf{k})$. For $\epsilon > 0$ and $h = 0$, $<\tilde{\phi}(\mathbf{x})> = 0$. We have

$$S(\tilde{k}) \propto \frac{\epsilon}{R^d \epsilon^{2-d/2}} \frac{1}{\tilde{k}^2 + 1}. \tag{15}$$

The Fourier transform of Eq. (15) gives the pair distribution function, which we write in terms of unscaled variables:

$$\rho^{(2)}(x) \sim \frac{1}{(x/\xi)^{d-2}} \frac{\epsilon}{R^d \epsilon^{2-d/2}} e^{-x/\xi}. \tag{16}$$

The $(x/\xi)^{2-d}$ dependence in Eq. (16) is valid for $d \geq 3$. For $d = 2$ this dependence is replaced by $(x/\xi)^{-1/2}$; in $d = 1$ there is no x-dependence in the denominator. The integral $\int d\mathbf{x}\, \rho^{(2)}(x)$ is proportional to $(\epsilon/R^d\epsilon^{2-d/2})\xi^d$ for all d. For scaling purposes we can treat $\rho^{(2)}(\mathbf{x})$ for $x \lesssim \xi$ as a constant. Because $\rho^{(2)}(\mathbf{x} \lesssim \xi)$ is proportional to the square of the density of a fluctuation, we see that the fluctuations in the order parameter density scale as

$$\phi_{\rm f}(\mathbf{x} \lesssim \xi) \sim \frac{\epsilon^{1/2}}{(R^d\epsilon^{2-d/2})^{1/2}} = \frac{\epsilon^{1/2}}{G^{1/2}}. \tag{17}$$

Note that the density of a critical phenomena fluctuation does not scale as $\epsilon^{1/2}$ as might be expected from a simple extension of how the order parameter scales for $\epsilon < 0$ in Eq. (9b). We will discuss this point more fully in Sec. IV. The scaling of $\phi_{\rm f}(\mathbf{x})$ with $G^{-1/2}$ in Eq. (17) justifies the neglect of the ϕ^4 term in Eq. (12) and the Gaussian approximation in Eq. (13).

The susceptibility χ is related to the pair distribution function $\rho^{(2)}$ by [32, 33]

$$\chi \propto \int d\mathbf{x}\, [\rho^{(2)}(\mathbf{x}) - \phi^2], \tag{18}$$

If we use the scaling form (17) of $\phi_{\rm f}(\mathbf{x})$ in Eq. (18), that is, $\chi \sim \phi_{\rm f}(\mathbf{x})^2\xi^d$, we find $\chi \sim \epsilon^{-1}$, consistent with Eq. (9c).

To show that $\phi_{\rm f}$ and ϕ have similar scaling behavior in a system with $R \sim 1$ and $d < 4$, we again assume that $\rho^{(2)}(\mathbf{x} \lesssim \xi)$ is a constant so that we can write Eq. (18) as $\chi \sim \phi_f^2\xi^d$ and $\phi_f^2 \sim \epsilon^{-\gamma}\epsilon^{d\nu}$. Hyperscaling [32, 33] gives $\gamma + 2\beta = d\nu$ so that $\phi_f^2 \sim \epsilon^{2\beta}$ and hence $\phi_f \sim \epsilon^{\beta}$.

We next discuss the Landau-Ginzburg and Cahn-Hilliard-Cook equations in (near-)mean-field systems. We can obtain these equations by noting that the time rate of change of an order parameter such as the density is related to the chemical potential μ. If the order parameter is not conserved and there are no other conservation laws (model A in the Hohenberg-Halperin classification scheme [34]), then

$$\frac{\partial\phi(t)}{\partial t} \propto -\mu \text{ and } \mu \propto \frac{\delta F(\phi)}{\delta\phi}, \tag{19}$$

where $F(\phi)$ is the free energy. We take $F(\phi)$ to be equal to the Landau-Ginzburg-Wilson Hamiltonian in Eq. (4), which is correct for mean-field systems and a good approximation in near-mean-field systems, and assume that the relations in Eq. (19) are valid in a spatial and time dependent context and that the functional derivatives are with respect to $\phi(\mathbf{x}, t)$. In this way we obtain the Landau-Ginzburg equation [19, 28]:

$$\frac{\partial\phi(\mathbf{x}, t)}{\partial t} = -M_A[-R^2\nabla^2\phi(\mathbf{x}, t) + 2\epsilon\phi(\mathbf{x}, t) + 4\phi^3(\mathbf{x}, t) - h] + \eta(\mathbf{x}, t), \tag{20}$$

where we have added a noise term $\eta(\mathbf{x},t)$. In the remainder of this section ϵ can be either positive or negative. For a conserved order parameter (model B [34]) we have

$$\frac{\partial\phi(\mathbf{x},t)}{\partial t} \propto \nabla\cdot\mathbf{J} \text{ and } \mathbf{J}\propto\nabla\mu(\mathbf{x},t). \tag{21}$$

If we again interpret the right-hand side of Eq. (4) as a free energy and include a noise term, we obtain the Cahn-Hilliard-Cook equation [19]

$$\frac{\partial\phi(\mathbf{x},t)}{\partial t} = M_B\nabla^2[-R^2\nabla^2\phi(\mathbf{x},t)+2\epsilon\phi(\mathbf{x},t)+4\phi^3(\mathbf{x},t)-h]+\eta_c(\mathbf{x},t). \tag{22}$$

The quantities M_A and M_B in Eqs. (20) and (22) are mobilities and will be discussed in Sec. III.

To obtain Eqs. (19) and (21) we assumed local equilibrium; that is, within the coarse grained volume used to obtain the Landau-Ginzburg free energy [19], the system comes into equilibrium on a time scale short compared to the time scales of interest.

For the remainder of this paper we will take $\eta(\mathbf{x},t)$ and $\eta_c(\mathbf{x},t)$ to be generated by a Gaussian distribution with zero mean. That is, $<\eta(\mathbf{x},t)> = <\eta_c(\mathbf{x},t)> = 0$, and

$$<\eta(\mathbf{x},t)\eta(\mathbf{x}',t')> = k_BT\delta(\mathbf{x}-\mathbf{x}')\delta(t-t') \tag{23a}$$

$$<\eta_c(\mathbf{x},t)\eta_c(\mathbf{x}',t')> = k_BT\nabla^2\delta(\mathbf{x}-\mathbf{x}')\delta(t-t'). \tag{23b}$$

We can use Eqs. (20) and (22) to determine the time dependence of the decay of fluctuations in (near-)mean-field systems. The scaling of $\phi_{\rm f}(\mathbf{x})$ in Eq. (17) implies that the cubic term in Eqs. (20) and (21) can be neglected. A straightforward calculation shows that the fluctuations decay exponentially with characteristic times that diverge as ϵ^{-1} in model A [32] and $R^2\epsilon^{-2}$ in model B [32]. For the remainder of this paper we will consider only model A.

III. PARISI-SOURLAS AND LIFETIME OF FLUCTUATIONS

The Parisi-Sourlas approach [29, 30] begins with the Landau-Ginzburg equation. Because the noise $\eta(\mathbf{x},t)$ in Eq. (20) is Gaussian, the measure of the noise is [29, 30]

$$P(\eta) = \frac{\exp[-\beta\int d\mathbf{x}dt\,\eta^2(\mathbf{x},t)]}{\int\delta\eta\exp[-\beta\int d\mathbf{x}dt\,\eta^2(\mathbf{x},t)]}. \tag{24}$$

We use Eq. (20) to replace $\eta(\mathbf{x},t)$, let $h=0$ for simplicity, and express Eq. (24) as

$$P(\phi)\propto J(\phi,\eta)\exp\Big\{-\beta\int d\mathbf{x}dt\Big[\frac{\partial\phi(\mathbf{x},t)}{\partial t}+M_A\big(-R^2\nabla^2\phi(\mathbf{x},t)+2\epsilon\phi(\mathbf{x},t)+4\phi^3(\mathbf{x},t)\big)\Big]^2\Big\}, \tag{25}$$

where the Jacobian $J(\phi,\eta)$ of the transformation from η to ϕ is the determinant of the operator $\delta\eta(\mathbf{x},t)/\delta\phi(\mathbf{x},t)$. Following Parisi and Sourlas [29, 30] we introduce the Grassman variables $\psi_F(\mathbf{x},t)$ and $\bar{\psi}_F(\mathbf{x},t)$ which satisfy the algebra

$$\psi_F^2(\mathbf{x},t) = \bar{\psi}_F^2(\mathbf{x},t) = \int d\psi_F(\mathbf{x},t) = \int d\bar{\psi}_F(\mathbf{x},t) = 0, \tag{26}$$

$$\big\{\bar{\psi}_F(\mathbf{x},t)\psi_F(\mathbf{x},t) + \psi_F(\mathbf{x},t)\bar{\psi}_F(\mathbf{x},t)\big\} = 0, \tag{27}$$

$$\int \psi_F(\mathbf{x},t) d\psi_F(\mathbf{x},t) = \int \bar{\psi}_F(\mathbf{x},t) d\bar{\psi}_F(\mathbf{x},t) = 1. \tag{28}$$

Because the variables ψ_F and $\bar{\psi}_F$ anticommute, they are referred to as as fermions; $\phi(\mathbf{x},t)$ is a boson. With this algebra we can evaluate the Jacobian in Eq. (25) and write $P(\phi)$ as

$$P(\phi,\psi_F,\bar{\psi}_F) = \frac{\exp\Big\{-\beta\big[\int d\mathbf{x}dt\,(S_B(\phi,\psi_F,\bar{\psi}_F) + S_F(\phi,\psi_F,\bar{\psi}_F)\big]\Big\}}{\bar{Z}}, \tag{29}$$

where $\bar{Z}$ is a normalization factor. The quantities S_B and S_F are given by

$$S_B(\phi,\psi_F,\bar{\psi}_F) = \int d\mathbf{x}dt\Big[\frac{\partial\phi(\mathbf{x},t)}{\partial t} + M_A\big(-R^2\nabla^2\phi(\mathbf{x},t) + 2\epsilon\phi(\mathbf{x},t) + 4\phi^3(\mathbf{x},t)\big)\Big]^2 \tag{30}$$

$$S_F(\phi,\psi_F,\bar{\psi}_F) = \int d\mathbf{x}dt\,\bar{\psi}_F(\mathbf{x},t)\Big[\frac{\partial}{\partial t} + M_A\big(-R^2\nabla^2 + 2\epsilon + 12\phi^2(\mathbf{x},t)\big)\Big]\psi_F(\mathbf{x},t). \tag{31}$$

We first consider $S_B(\phi,\psi_F,\bar{\psi}_F)$ in Eq. (30). Among the terms found by evaluating the square of the term in brackets is the contribution

$$C(\phi) = 2M_A\int d\mathbf{x}dt\frac{\partial\phi(\mathbf{x},t)}{\partial t}\big[-R^2\nabla^2\phi(\mathbf{x},t) + 2\epsilon\phi(\mathbf{x},t) + 4\phi^3(\mathbf{x},t)\big], \tag{32a}$$

$$= 2M_A\int d\mathbf{x}\int_{t_I}^{t_F} dt\frac{\partial}{\partial t}H(\phi(\mathbf{x},t)), \tag{32b}$$

where H is given by Eq. (4) with $h = 0$ and ϕ is replaced by $\phi(\mathbf{x},t)$. The integral with respect to t gives

$$C(\phi) = 2M_A\int d\mathbf{x}\big[H\big(\phi(\mathbf{x},t_F)\big) - H\big(\phi(\mathbf{x},t_I)\big)\big]. \tag{33}$$

Parisi and Sourlas assume that t_F and t_I can be found such that $C(\phi) = 0$ and show that with this assumption there is a transformation that maps fermions and bosons into each other and keeps $P(\phi)$ in Eq. (29) invariant. They refer to such systems as supersymmetric. If a system is in equilibrium, such values of t_I and t_F can always be found [29, 30].

The supersymmetric form of $P(\phi,\psi_F,\psi_F)$ is the proper representation for investigating the morphology of the fluctuations in the neighborhood of (near-)mean-field critical points.

Near the latter the Hamiltonian in Eq. (4) can be assumed to be Gaussian. This assumption implies that the Landau-Ginzburg and Cahn-Hilliard-Cook equations can be linearized for $G \gg 1$. Because the ϕ dependence in the fermionic contribution to the action $S_F(\phi, \psi_F, \bar{\psi}_F)$ comes from the nonlinear term in Eq. (20), linearization makes the two contributions to the action, $S_B(\phi, \psi_F, \bar{\psi}_F)$ and $S_F(\phi, \psi_F, \bar{\psi}_F)$, independent. The integration over the fermionic variables $\psi_F(\mathbf{x}, t)$ and $\bar{\psi}_F(\mathbf{x}, t)$ can be done immediately resulting in the measure

$$P(\phi) = \frac{\exp\left\{-\beta \int d\mathbf{x}dt \left(\frac{\partial\phi(\mathbf{x},t)}{\partial t}\right)^2 + M_A^2[-R^2\nabla^2\phi(\mathbf{x},t) + \epsilon\phi(\mathbf{x},t)]^2\right\}}{Z_S}, \tag{34}$$

where Z_S is the functional integral over ϕ of the numerator in Eq. (34).

In equilibrium $P(\phi_{\rm f})$ should give the same probability of a fluctuation as $P_B(\phi_{\rm f})$ in Eq. (6). To understand the relation between these two probabilities we note that if $P_B(\phi_{\rm f})$ is of order e^{-1}, then $P(\phi_{\rm f})$ should also be of order e^{-1}. (This requirement follows from the fact that we expect the probability of variations from equilibrium to decay exponentially.) If we take $\phi_{\rm f}$ in Eq. (34) to describe an equilibrium fluctuation, we expect that

$$\int d\mathbf{x}dt \Big(\frac{\partial\phi_{\rm f}(\mathbf{x},t)}{\partial t}\Big)^2 \sim A. \tag{35}$$

where A is a constant independent of $\phi_{\rm f}$. Without loss of generality we can set $A = 1$. Because the spatial extent of $\phi_{\rm f}$ scales as the correlation length ξ, we have from simple scaling arguments that $d\mathbf{x}$ in Eq. (35) scales as ξ^d. If we use the scaling of $\phi_{\rm f}(\mathbf{x})$ in Eq. (17) and the scaling of ξ in Eq. (9a), Eq. (35) implies

$$\frac{\phi_{\rm f}^2\,\xi^d}{\tau_{\rm f,c}} \sim \frac{\epsilon\, R^d\epsilon^{-d/2}}{R^d\epsilon^{2-d/2}\,\tau_{\rm f,c}} \sim 1, \tag{36}$$

or

$$\tau_{\rm f,c} \sim \epsilon^{-1}. \qquad \text{(lifetime of fluctuations)} \tag{37}$$

Equation (37) is the well known scaling relation for critical slowing down near mean-field critical points for model A [34].

If we require that $\int d\mathbf{x}dt\, M_A^2\big[-R^2\nabla^2\phi_{\rm f}(\mathbf{x},t) + \epsilon\phi_{\rm f}(\mathbf{x},t)\big]^2 \sim 1$ (see Eq. (34)), and use the scaling relations for ξ, $\phi_{\rm f}$, and $\tau_{\rm f,c}$ and the same arguments used to obtain Eq. (36), we find

$$\frac{M_A^2\epsilon^3 R^d\epsilon^{-d/2}\epsilon^{-1}}{R^d\epsilon^{2-d/2}} \sim 1, \tag{38}$$

which implies that M_A is a constant of order 1.

These results are all expected. Note that there is a significant conceptual difference between $P_B(\phi)$ and $P(\phi)$. The quantity $P_B(\phi)$ is the fraction of independent members of an ensemble in which $\phi(\mathbf{x})$ is realized when a measurement is made. Because the system is in equilibrium, we can divide a time sequence of measurements into independent segments that can be thought of as members of an ensemble. These segments have a duration of the order of the decorrelation time (or longer), which is of order $\tau_{\rm f,c}$ near the critical point. The quantity $P(\phi)$ gives the probability of a "path" in ϕ space. The paths of interest here are those whose probability is the order of e^{-1}. The path that results in an object with a density difference from the background of magnitude $\phi_{\rm f} = \epsilon^{1/2}/G^{1/2}$, spatial extent of order $\xi = R\epsilon^{-1/2}$, and a lifetime of order ϵ^{-1} is one such path.

Suppose that in equilibrium there is an object with spatial extent $\xi = R\epsilon^{-1/2}$ but a different density. In particular, suppose there is an object of density of $\phi_{\rm fc} \sim \epsilon^{1/2}/G$. For reasons that we will discuss in Sec. V we will call this object a fundamental cluster. Because we have assumed equilibrium, we have supersymmetry, and the action is the sum of two contributions as in Eq. (34). One term has the form as in Eq. (35):

$$S_{B,\,1} = \int d\mathbf{x}dt\Big(\frac{\partial\phi(\mathbf{x},t)}{\partial t}\Big)^2. \tag{39}$$

We use reasoning similar to that following Eq. (35) and define the lifetime of the fundamental cluster to be given by $S_{B,\,1} \sim 1$. If we substitute $\phi_{\rm fc} \sim \epsilon^{1/2}/G$ in Eq. (39), we obtain

$$\frac{\phi_{\rm fc}^2\,\xi^d}{\tau_{\rm fc,c}} \sim \frac{\epsilon R^d \epsilon^{-d/2}}{(R^d\epsilon^{2-d/2})^2\,\tau_{\rm fc,c}} \sim 1, \tag{40}$$

or

$$\tau_{\rm fc,c} \sim \frac{\epsilon^{-1}}{R^d\epsilon^{2-d/2}} = \frac{\epsilon^{-1}}{G}. \qquad \text{(lifetime of fundamental cluster)} \tag{41}$$

Equation (41) gives the lifetime of an object (the fundamental cluster) with density difference from the background $\phi_{\rm fc} \sim \epsilon^{1/2}/G$ near a mean-field critical point.

The Boltzmann probability $P_B(\phi)$ of finding such an object is

$$P_B(\phi_{\rm fc}) \propto \exp\Big\{-\beta\int d\mathbf{x}\Big[\frac{R^2}{2}[\nabla\phi_{\rm fc}(\mathbf{x})]^2 + \epsilon{\phi_{\rm fc}}^2(\mathbf{x})\Big]\Big\}. \tag{42}$$

If we use the scaling relations and set $\beta = 1$ for convenience, we obtain

$$P_B(\phi_{\rm fc}) \propto e^{-1/R^d\epsilon^{2-d/2}}. \tag{43}$$

We see that $P_B(\phi_{\rm fc}) \neq P(\phi_{\rm fc})$ despite the fact that the system is in equilibrium. If we want the probability $P(\phi_{\rm fc})$ that there is a path that consists of an object (the fundamental

cluster) with density $\phi_{\rm fc} \sim \epsilon^{1/2}/G$, spatial extent $\xi \sim R\epsilon^{-1/2}$, and lifetime ϵ^{-1} rather than ϵ^{-1}/G, then using Eq. (34) we have

$$P(\phi_{\rm fc}) \sim e^{-1/R^d\epsilon^{2-d/2}}, \tag{44}$$

and $P_B(\phi_{\rm fc}) = P(\phi_{\rm fc})$. The implication of these results is that the Boltzmann probability $P_B(\phi_{\rm fc})$ requires a given time, that is, the decorrelation time, which near a mean-field critical point scales as ϵ^{-1}. The probability $P(\phi_{\rm fc}) = P_B(\phi_{\rm fc})$ only if t is chosen to be the decorrelation time $\tau_{\rm f,c}$. In general, the decorrelation time is not equal to the lifetime of the object of interest. Note that the same arguments apply to the normalization factors Z in Eq. (12) and Z_S in Eq. (34). In particular, $Z = Z_S$ only if the time scale is chosen to be $\tau \sim \epsilon^{-1}$.

The mobility M_A need not be a constant independent of ϵ [19]. For an object with density $\phi_{\rm fc} \sim \epsilon^{1/2}/G$, the second term in the action in Eq. (25) has the form

$$S_2 = -\int d\mathbf{x}dt\, {M_A}^2\big[R^2[\nabla\phi_{\rm fc}(\mathbf{x},t)]^2 + 2\epsilon\phi_{\rm fc}(\mathbf{x},t)\big]^2. \tag{45}$$

If we use the scaling relations and the lifetime given by Eq. (41), we obtain

$$\frac{{M_A}^2\epsilon^3 R^d\epsilon^{-d/2}\epsilon^{-1}}{(R^d\epsilon^{2-d/2})^3} \sim 1, \tag{46}$$

or

$$M_A \sim R^d\epsilon^{2-d/2} \gg 1. \tag{47}$$

M_A in Eq. (47) depends on ϵ in contrast to the mobility in Eq. (38). If we had considered a lifetime of ϵ^{-1} rather than ϵ^{-1}/G in Eq. (41), M_A would be order unity.

In summary, the probabilities $P_B(\phi)$ and $P(\phi)$ are equal if the lifetime of an object is the order of ϵ^{-1} near the mean-field critical point. The lifetime of an object is obtained by requiring that $P(\phi) \sim e^{-1}$. For objects with density $\epsilon^{1/2}/G$ and a lifetime of ϵ^{-1}/G, the usual Boltzmann factor will not give the probability of observing such an object.

IV. SPINODALS AND PSEUDOSPINODALS

In this section we discuss the meaning of spinodals and pseudospinodals. We begin with the Hamiltonian in Eq. (4) and the partition function in Eq. (5). For $G \gg 1$ the partition function can be evaluated using saddle point techniques, and the free energy has

the Landau-Ginzburg form [28],

$$F = \int d\mathbf{x} \Big[\frac{R^2}{2} [\nabla \phi(\mathbf{x})]^2 + \epsilon \phi^2(\mathbf{x}) + \phi^4(\mathbf{x}) - h\phi(\mathbf{x}) \Big]. \tag{48}$$

We set the gradient term equal to zero to obtain the free energy density

$$f = \epsilon \phi^2 + \phi^4 - h\phi. \tag{49}$$

For $\epsilon > 0$ there is only one real extremum of the free energy. For $\epsilon < 0$ there are three real extrema, one maximum and two minima. For $h = 0$ there are two states or values of ϕ with the same free energy. As $|h|$ is increased, one of the minima becomes higher than the other. The higher minimum corresponds to the metastable state. Increasing $|h|$ further eventually results in the disappearance of the metastable minimum. This value of $|h|$ is referred to as the spinodal field h_s. It is easy to see from Eq. (49) that at $h = h_s$, f has an inflection point at $\phi = \phi_s$. If we set (for $\epsilon < 0$) $\partial f/\partial \phi = \partial^2 f/\partial \phi^2 = 0$, we find

$$\phi_s = (|\epsilon|/6)^{1/2} \text{ and } h_s = 4|\epsilon|^{3/2}/(3\sqrt{6}). \tag{50}$$

We define the new variable Δh,

$$\Delta h = h_s - h, \tag{51}$$

and the new field, $\psi(\mathbf{x}) = \phi(\mathbf{x}) - \phi_s + a$, and write the mean-field free energy as

$$F = \int d\mathbf{x} \Big[\frac{R^2}{2} [\nabla \psi(\mathbf{x})]^2 + \Delta h^{1/2} \lambda_1 \psi^2(\mathbf{x}) - \lambda_2 \psi^3(\mathbf{x}) + \lambda_3 \psi^4(\mathbf{x}) \Big]. \tag{52}$$

The parameter a is chosen so that the term linear in $\psi(\mathbf{x})$ does not appear in F. The coefficients λ_i are functions of ϵ and independent of Δh. (More precisely, the λ_i are a function of Δh, but as $\Delta h \to 0$, the λ_i approach constants.) The free energy in Eq. (52) is constructed so that the spinodal is at $\psi = 0$ and $\Delta h = 0$.

As for the critical point we assume that the fluctuations associated with the spinodal can be described by a Gaussian-Landau-Ginzburg-Wilson Hamiltonian with the partition function

$$Z = \int \delta\psi \exp \big[-\beta \int d\mathbf{x} \frac{R^2}{2} [\nabla \psi(\mathbf{x})]^2 + \Delta h^{1/2} \lambda_1 \psi^2(\mathbf{x}) \big]. \tag{53}$$

If we follow the same argument that we used at the critical point in Sec. II, we obtain

$$\xi \sim R \Delta h^{-1/4} \tag{54a}$$

$$\chi \sim \Delta h^{-1/2}. \tag{54b}$$

The fluctuations of the order parameter density scale as

$$\psi_f(\mathbf{x} \lesssim \xi) \sim \frac{\Delta h^{1/2}}{[R^d \Delta h^{3/2-d/4}]^{1/2}} = \frac{\Delta h^{1/2}}{G_s^{1/2}}, \tag{54c}$$

where the Ginzburg parameter near the spinodal is

$$G_s = R^d \Delta h^{3/2-d/4}. \tag{55}$$

The system is mean-field when $G_s \to \infty$ and near-mean-field for $G_s \gg 1$. A spinodal is only a true critical point in the mean-field limit [7, 8, 9]. The distinction between a spinodal ($G_s \to \infty$) and a pseudospinodal ($G_s \gg 1$) will be clear from the context.

As discussed in Sec. II the scaling of the order parameter density and the density of the order parameter fluctuations is not the same in (near-)mean-field systems. For example, near the critical point for $\epsilon < 0$ we have from Eq. (49)

$$-2|\epsilon|\phi + 4\phi^3 = 0. \tag{56}$$

Equation (56) gives the order parameter density at the minima for $\epsilon < 0$. As $|\epsilon| \to 0$ we have

$$\phi \sim |\epsilon|^{1/2}. \tag{57}$$

Near the spinodal we use Eq. (52) and assume that $\psi(\mathbf{x})$ is independent of $\mathbf{x}$. We have

$$f(\psi) = \Delta h^{1/2} \lambda_1 \psi^2 - \lambda_2 \psi^3 + \lambda_3 \psi^4. \tag{58}$$

As $\Delta h \to 0$, ψ scales as

$$\psi \sim \Delta h^{1/2} \qquad \text{(order parameter density near the spinodal)}, \tag{59}$$

where we have dropped the ψ^4 term in Eq. (58). Equation (59) gives the scaling of the order parameter density. As near the critical point, the scaling of the order parameter density and the density of the fluctuations is not the same in mean-field systems near the spinodal.

In the above discussion we kept the temperature fixed and approached the spinodal by varying the magnetic field h. Alternatively, we can keep the magnetic field fixed and approach the spinodal by varying the temperature. To obtain the critical exponents in the temperature variable we return to Eq. (49) and write ϵ as

$$\epsilon = \frac{T - T_c}{T_c} = \frac{T - T_s}{T_c} + \frac{T_s - T_c}{T_c} = \varepsilon + \Delta_s, \tag{60}$$

where T_s is the spinodal temperature for a fixed field $h = h_s$. We write

$$\frac{\partial^2 f}{\partial \phi^2}\Big|_{\phi=\phi_s} = -2\varepsilon - 2|\Delta_s| + 12\phi_s^2 + 24\phi_s\psi = 0, \tag{61}$$

Because Δ_s and ϕ_s are on the spinodal curve, we have

$$-2|\Delta_s| + 12\phi_s^2 = 0, \tag{62}$$

$$-2|\varepsilon| + 24\phi_s\psi = 0. \tag{63}$$

From Eqs. (59) and (63) we have

$$\Delta h^{1/2} \sim \varepsilon, \tag{64}$$

which implies from Eq. (54c) that as the spinodal is approached, the density of the order parameter fluctuations scales as

$$\psi_f(\mathbf{x} \lesssim \xi) \sim \frac{\varepsilon}{[R^d \varepsilon^{3-d/2}]^{1/2}}. \tag{65a}$$

Similarly, the correlation length scales as

$$\xi \sim R\varepsilon^{-1/2}, \tag{65b}$$

and the suceptibility diverges as

$$\chi \sim \varepsilon^{-1}. \tag{65c}$$

Equations (54) and (65) give the critical exponents near the spinodal in terms of Δh and ε.

We first discuss the application of the Parisi-Sourlas method near the spinodal. If we construct a Landau-Ginzburg equation from the free energy in Eq. (52), we have

$$\frac{\partial \psi(\mathbf{x},t)}{\partial t} = -M_{A,s}\big[-R^2\nabla^2\psi(\mathbf{x},t) + 2\Delta h^{1/2}\lambda_1\psi(\mathbf{x},t) - 3\lambda_2\psi^2(\mathbf{x},t) + 4\lambda_3\psi^3(\mathbf{x},t)\big] + \eta(\mathbf{x},t). \tag{66}$$

Because the noise $\eta(\mathbf{x},t)$ is Gaussian, we obtain an expression for the probability of a path $\psi(\mathbf{x},t)$ of the form

$$P_{\rm sp}(\psi) = \frac{\exp\big[-\beta\int d\mathbf{x}dt\,\big(\frac{\partial\psi(\mathbf{x},t)}{\partial t}\big)^2 + M_A^2\big\{-R^2\nabla^2\psi(\mathbf{x},t) + 2\lambda_1\Delta h^{1/2}\psi(\mathbf{x},t)\big\}^2\big]}{Z_{\rm sp}}, \tag{67}$$

where we have used the linear form of the Landau-Ginzburg equation and have assumed that the system is in metastable equilibrium, which implies supersymmetry. Arguments similar to those used at the critical point show that the relaxation or decorrelation time scales as

$$\tau \sim \Delta h^{-1/2}. \qquad \text{(decorrelation time near the spinodal)} \tag{68}$$

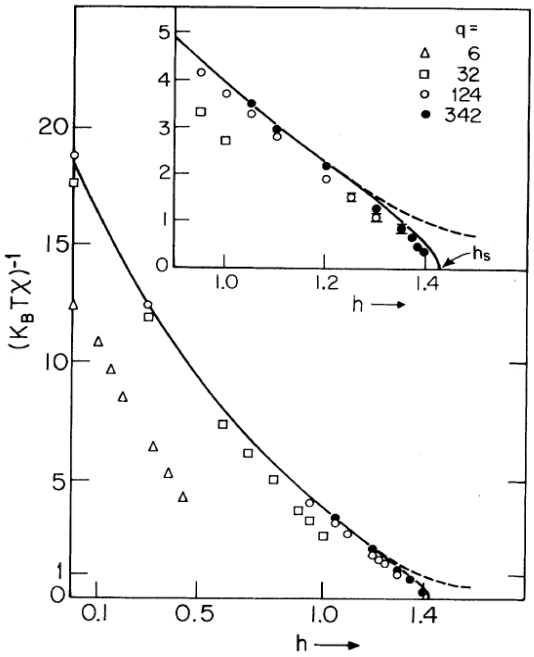

FIG. 1: The inverse susceptibility as a function of the magnetic field h (from Ref. 7). Note that as the number of neighbors q is increased, the inverse susceptibility more closely follows a power law. The inset shows the behavior of $(k_B T\chi)^{-1}$ closer to the pseudospinodal.

All considerations of the difference between equilibrium Boltzmann probabilities and probabilities of paths are the same near spinodals and critical points.

We now consider the nature of the pseudospinodal. As mentioned, for $G_s \gg 1$ but finite, there is no spinodal. However, the system behaves as if it existed if G_s is sufficiently large. In Fig. 1 we plot the inverse of the isothermal susceptibility χ found by a Monte Carlo simulation for a $d = 3$ Ising model as a function of the applied magnetic field h for different values of R [7]. The temperature is taken to be $4T_c/9$, where T_c is the critical point temperature. The solid line is the mean-field prediction for $q \to \infty$, where q is the number of spins that interact with a given spin [35]. Data was taken only if the metastable state lived longer than 10^4 Monte Carlo time steps per spin. For nearest-neighbor interactions ($q = 6$) the data stops at $h \sim 0.5$ far from the spinodal value of the field $h_s = 1.43$. As R and hence q is increased, the data approaches the mean-field result and the spinodal can be more closely approached. This result indicates that the larger the value of R, the more system behaves like there is an underlying spinodal.

Another way to understand the nature of the pseudospinodal is to look at the behavior

of the zeros of the partition function as a function of R. The zeros of the partition function corresponding to the spinodal lie in the four-dimensional complex magnetic field-temperature space for finite R [9]. As R is increased, the zeros move toward the real (h, T) plane similar to the behavior of the zeros of the partition function for Ising models in finite systems as the system size increases [36, 37]. The idea is that the pseudospinodal appears to be a critical point if h is not too close to h_s. What is meant by too close can be estimated by the magnitude of the Ginzburg parameter G_s in Eq. (55). The value of Δh where the spinodal concept fails can be made smaller by increasing R. Hence, the theoretical arguments we made about the properties of fluctuations near the spinodal can be tested in systems where the interaction range R is large, even though there is no true spinodal in nature. However, such statements have to be modified in systems with a phase transition that involve spatial symmetry breaking such as the liquid-solid transition (see Sec. VII).

V. FLUCTUATION STRUCTURE

In this section we will use scaling arguments and the cluster mapping discussed in Appendix A to determine the structure of the fluctuations in (near-)mean-field systems.

Ising critical points and the spinodal in (near-)mean-field systems have been mapped onto percolation transitions [25, 38]. For simplicity, we will assume that the interaction between spins in is a constant up to a distance R and is zero for distances greater than R. To map the critical point onto a percolation transition we toss bonds randomly between pairs of parallel spins that are separated by a distance less than or equal to R with a probability $p_b = 1 - e^{-2\beta J}$ where J is the usual Ising coupling constant. This mapping guarantees that the percolation transition occurs at the critical point. For the spinodal the bond probability is $p_s = 1 - e^{-\beta J(1-\rho)}$, where ρ is the density of spins in the stable state direction; that is, if the metastable state is in the up direction, then ρ is the density of spins in the down direction. The size of the clusters is determined by the number of spins in a cluster. The details of this mapping are given in Appendix A.

Equilibrium critical phenomena fluctuations in the order parameter are defined as deviations from its mean value with a linear dimension of the correlation length and a free energy cost of order 1. These properties were used in Sec. II to calculate the density of fluctuations near a critical point. We will see that the same properties apply near a spinodal.

The geometrical quantity that is isomorphic to the free energy is $-k_BT$ times the mean number of clusters (see Appendix A). The free energy density near the critical point scales as ϵ^2, because the specific heat exponent $\alpha = 0$ for the mean-field critical point [32, 33]. Hence the free energy $F(\epsilon)$ in a correlation length volume scales as

$$F(\epsilon) \sim \epsilon^2 \xi^d = R^d \epsilon^{2-d/2}, \tag{69}$$

and the mean number of clusters in a correlation length volume scales as

$$\overline{n}_{\rm fc,c} \sim R^d \epsilon^{2-d/2} = G. \tag{70}$$

How are the clusters related to the fluctuations? In non-mean-field systems such as the Ising model with $R \sim 1$ and $d < 4$, the mean number of clusters in a correlation length volume near the critical point scales as $\epsilon^{2-\alpha}\xi^d \sim 1$. The isomorphism between the Ising model and percolation implies that the the pair distribution function $\rho^{(2)}$ is the same as the pair connectedness function $\rho_{\rm c}^{(2)}$, which is the probability that two spins a distance x apart belong to the same cluster. For $x \lesssim \xi$, $\rho_{\rm c}^{(2)}$ is roughly a constant and equal to $\phi_{\rm cl}^2$, where $\phi_{\rm cl}$ is the density of spins in the cluster. This density must be equal to the density of a fluctuation which scales as ϵ^β (see Eq. (9b)). Hence, $\phi_{\rm cl}$ scales as ϵ^β in a non-mean-field system, and the clusters are a statistical realization of the fluctuations [38, 39].

For $G \gg 1$ the number of clusters in a correlation length volume near the critical point scales as G (see Eq. (70)), which is much larger than unity. Are the clusters a statistical realization of the fluctuations? To understand that the answer is no, we note from Eq. (17) that the density of a critical phenomena fluctuation scales as $\phi_{\rm f}(\mathbf{x}) \sim \epsilon^{1/2}/(R^d\epsilon^{2-d/2})^{1/2}$. If a single cluster were to correspond to a fluctuation, then the density of spins in a correlation length volume would scale as

$$G\phi_{\rm f}(\mathbf{x}) = R^d\epsilon^{2-d/2}\frac{\epsilon^{1/2}}{[R^d\epsilon^{2-d/2}]^{1/2}} = [R^d\epsilon^{2-d/2}]^{1/2}\epsilon^{1/2}. \tag{71}$$

Because the mean-field limit corresponds to letting $R \to \infty$ before $\epsilon \to 0$ [5], Eq. (71) implies that the spin density is infinite in the mean-field limit, which is impossible. Hence, the density of the clusters must be much smaller than the density of the fluctuations, and a fluctuation does not correspond to a single cluster as it does in short-range systems. We conclude that the clusters in (near-)mean-field systems play a different role, and we will refer to the clusters in these systems as *fundamental clusters*.

To understand the relation between the fundamental clusters and the fluctuations in a (near-)mean-field system, we again use the the fact that the pair distribution function $\rho^{(2)}$ is isomorphic to the pair connectedness function $\rho_c^{(2)}$. Because the latter is the probability that two sites a distance x apart belong to the same cluster, we have

$$\rho_c^{(2)}(\mathbf{x} \lesssim \xi) \sim p_{\rm fc,c} \frac{p_{\rm fc,c}}{R^d \epsilon^{2-d/2}}, \tag{72}$$

where $p_{\rm fc,c}$ is the probability that the first site belongs to any one of the $R^d\epsilon^{2-d/2}$ clusters, and $p_{\rm fc,c}/R^d\epsilon^{2-d/2}$ is equal to the cluster density, which is the probability that another site belongs to the same cluster as the first.

Similarly, we have that

$$\rho^{(2)}(\mathbf{x} \lesssim \xi) \sim \phi_f^2(\mathbf{x} \lesssim \xi) \sim \left[\frac{\epsilon^{1/2}}{(R^d\epsilon^{2-d/2})^{1/2}}\right]^2. \tag{73}$$

We have $\rho_c^{(2)}(\mathbf{x} \lesssim \xi) = \rho_c^{(2)}(\mathbf{x} \lesssim \xi)$, and hence We have

$$\frac{p_{\rm fc,c}^2}{R^d\epsilon^{2-d/2}} = \left[\frac{\epsilon^{1/2}}{\left(R^d\epsilon^{2-d/2}\right)^{1/2}}\right]^2, \tag{74}$$

and $p_{\rm fc,c} = \epsilon^{1/2}$. Hence, the density of spins in a fundamental cluster is

$$\phi_{\rm fc,c}(\mathbf{x} \lesssim \xi) \sim \frac{p_{\rm fc,c}}{R^d\epsilon^{2-d/2}} = \frac{\epsilon^{1/2}}{R^d\epsilon^{2-d/2}} = \frac{\epsilon^{1/2}}{G}. \tag{75}$$

Because the density of the fundamental clusters is much smaller than the density of the fluctuations for $G \gg 1$, a fluctuation must correspond to many fundamental clusters.

We can test the prediction for $\phi_{\rm fc,c}(\mathbf{x})$ in Eq. (75) by determining the dependence of $m_{\rm fc,c}$, the mean number of spins in a fundamental cluster, on ϵ. This dependence is given by

$$m_{\rm fc,c} \sim \phi_{\rm fc,c}\xi^d \sim \frac{\epsilon^{1/2}}{R^d\epsilon^{2-d/2}} R^d \epsilon^{-d/2} = \epsilon^{-3/2}. \tag{76}$$

In Fig. 2 we plot $m_{\rm cl}$ a function of ϵ for fixed R. The slope of the log-log plot is consistent with the theoretical prediction in Eq. (76).

We now discuss the relation between the fundamental clusters and the fluctuations in more detail. As discussed in Appendix A the clusters are constructed to be independent. Therefore a given cluster can "flip" independently of the other clusters. There are $R^d\epsilon^{2-d/2}$ clusters near the mean-field critical point, half up and half down by symmetry. Because

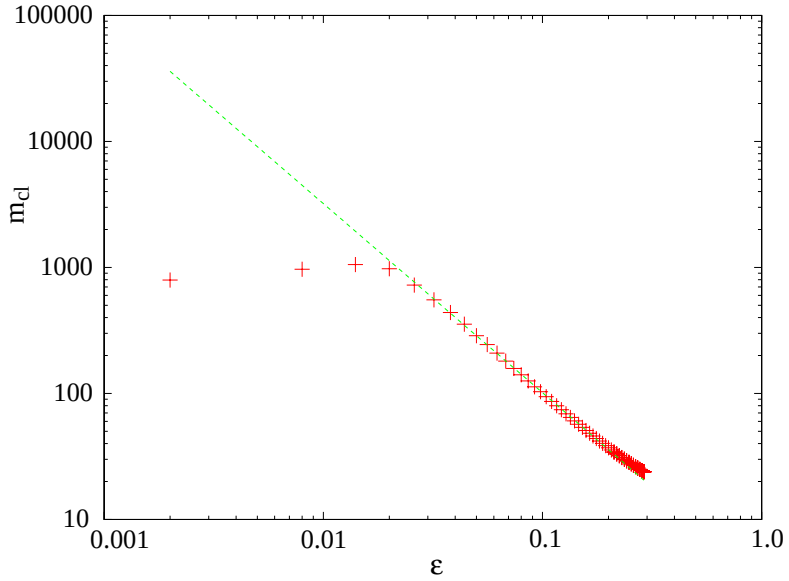

FIG. 2: The mean number of spins in a fundamental cluster with spatial extent ξ found in a Monte Carlo simulation of the $d = 2$ Ising model with $R = 20$ near T_c as a function of ϵ. The linear dimension of the system is $L = 240$. The slope is ≈ -1.5 if the data is fitted in the range $[0.03, 0.2]$. Note the deviation of $m_{\rm cl}$ from the mean-field prediction near the mean-field critical point where G is too small to apply mean-field arguments and where finite size effects become important.

the clusters are independent, the mean number of excess fundamental clusters *in a given direction* is determined by a random walk:

$$\overline{\Delta}_{\rm fc,\,c} \sim [R^d \epsilon^{2-d/2}]^{1/2} = G^{1/2}. \tag{77}$$

More precisely, the distribution of the number of fundamental clusters in a fluctuation is a Gaussian peaked about $\overline{\Delta}_{\rm fc,\,c}$.

From this analysis we see that the density of a fluctuation is the product of the density of fundamental clusters, Eq. (75), and the mean number of excess clusters, Eq. (77):

$$\phi_{\rm f}(\mathbf{x} \lesssim \xi) \sim \phi_{\rm fc,c}\overline{\Delta}_{\rm fc,\,c} \sim \frac{\epsilon^{1/2}}{R^d\epsilon^{2-d/2}}(R^d\epsilon^{2-d/2})^{1/2} = \frac{\epsilon^{1/2}}{(R^d\epsilon^{2-d/2})^{1/2}} = \frac{\epsilon^{1/2}}{G^{1/2}}, \tag{78}$$

in agreement with Eq. (17).

A similar analysis can be done near the spinodal, and we will only summarize the results here. In this case there is an infinite cluster that is a statistical realization of the metastable state magnetization [23, 24]. If we subtract this cluster [40], the results are similar to those near the mean-field critical point. The mean number of fundamental clusters in a correlation

length volume, half up and half down, is given by (compare to Eq. (70))

$$\overline{n}_{\mathrm{fc,\,s}} \sim R^d \Delta h^{3/2-d/4} = G_s, \tag{79}$$

and their density is (see Eq. (75))

$$\psi_{\mathrm{fc,s}} \sim \frac{\Delta h^{1/2}}{R^d \Delta h^{3/2-d/4}} = \frac{\Delta h^{1/2}}{G_s}. \tag{80}$$

We see that a system is near-mean-field when the number of fundamental clusters in a correlation length volume is large (see Eq. (79)). As $\epsilon\,(\Delta h)$ is decreased for $d < 4$ (critical point) or $d < 6$ (spinodal) for fixed R, $G\,(G_s)$ decreases and the system becomes less mean-field.

If we use the same random walk argument as for Eq. (77), we find that the density of the order parameter fluctuations near the spinodal scales as

$$\psi_{f,s}(\mathbf{x} \lesssim \xi) \sim \frac{\Delta h^{1/2}}{(R^d \Delta h^{3/2-d/4})^{1/2}} = \frac{\Delta h^{1/2}}{G_s^{1/2}}. \tag{81}$$

Hyperscaling (two exponent scaling) is satisfied for systems that are non-mean-field if ϵ (Δh) is sufficiently small [32, 33]. This connection suggests that the magnitude of $G\,(G_s)$ determines the existence of hyperscaling. If $G\,(G_s)$ decreases, the clusters must coalesce and additional length scales are introduced [41]. We now show that the existence of one relevant or divergent length scale is insufficient for the existence of hyperscaling if $G \gg 1$ because the number of fundamental clusters changes as the critical point is approached. The assumption of one divergent length scale leads to the following form for the singular part of the free energy density near the critical point [32, 33]

$$f(\epsilon, h) = \frac{1}{\xi^d} f(\xi^{y_T}\epsilon, \xi^{y_h} h). \tag{82}$$

If we differentiate f twice with respect to h and set $h = 0$, we find

$$\left.\frac{\partial^2 f(\epsilon, h)}{\partial h^2}\right|_{h=0} = \frac{\xi^{2y_h}}{\xi^d} \left.\frac{\partial^2}{\partial (\xi^{y_h})^2} f(\xi^{y_T}\epsilon, \xi^{y_h} h)\right|_{h=0}. \tag{83}$$

The left-hand side of Eq. (83) is the isothermal susceptibility χ. We now fix $\xi^{y_T}\epsilon$ to be equal to one. Because $f(1, h = 0)$ is not singular [32, 33] and $\xi \sim \epsilon^{-1/y_T}$, we have $\xi^{2y_h - d} = \epsilon^{-(2y_h - d)/y_T}$. Hence the exponent γ that characterizes the divergence of χ near the critical point is given by

$$\gamma = \frac{2y_h - d}{y_T}. \tag{84}$$

By using a similar argument, we obtain

$$\beta = \frac{d - y_h}{y_T}. \tag{85}$$

For mean-field Ising models or simple fluids $\beta = 1/2$ and $\gamma = 1$. Hence $y_h = 3d/4$ and $y_T = d/2$. For fixed R, $\xi = R\epsilon^{-1/2}$, so that $y_T = 2$ for all d [32, 33] and not $d/2$. Hence, for fixed R two exponent scaling does not hold in the neighborhood of a mean-field critical point. The same argument holds near the spinodal.

In contrast, consider what happens for fixed $G = R^d\epsilon^{2-d/2}$. From Eqs. (17) and (18) we have

$$\chi \sim \Big[\frac{\epsilon^{1/2}}{(R^d\epsilon^{2-d/2})^{1/2}}\Big]^2 R^d\epsilon^{-d/2}. \tag{86}$$

If we keep $G = R^d\epsilon^{2-d/2}$ constant, then $R^d\epsilon^{-d/2} \propto \epsilon^{-2}$. Therefore the susceptibility $\chi \propto \epsilon^{-1}$ so $\gamma = 1$. Likewise from Eq. (78) the density scales as $\epsilon^{1/2}$ so $\beta = 1/2$. For G fixed $R\epsilon^{-1/2} \propto \epsilon^{-2/d}$ so that $\nu = 2/d$ and $y_T = d/2$. Hyperscaling now holds and $\gamma + 2\beta = d\nu$. Similar arguments hold near the spinodal if the infinite cluster is removed.

In summary, we have shown that the relation between the clusters and the fluctuations in (near-)mean-field systems is more complex than in non-mean-field systems. In particular, the individual clusters are not realizations of the fluctuations, which instead are related to fluctuations of the number of clusters. For this reason we refer to the clusters in (near-)mean-field systems as fundamental clusters. The mean number of fundamental clusters in a correlation length volume is proportional to the Ginzburg parameter G. The dependence of G on ϵ (G_s on Δh) causes the breakdown of hyperscaling.

VI. LIFETIME OF THE FUNDAMENTAL CLUSTERS

In Sec. V we assumed a scaling form for the free energy. Here we do a more detailed calculation using the Landau-Ginzburg-Wilson Hamiltonian in the mean-field limit. Our main result is the scaling dependence of the lifetime of the fundamental clusters. We also recover the same scaling results for the free energy.

If we set $h = 0$, scale all lengths with the correlation length, and assume that $\phi(\mathbf{x})$ scales as $\epsilon^{1/2}$ near the critical point, we obtain the Hamiltonian in Eq. (8). If we assume that $\tilde{\phi}(\mathbf{y})$ is independent of $\mathbf{y}$ and restrict the integral to a region the size of a correlation length volume, we have

$$H(\phi) = R^d\epsilon^{2-d/2}[\pm\tilde{\phi}^2 + \tilde{\phi}^4], \tag{87}$$

and the partition function becomes

$$Z(\epsilon) = \int_{-\infty}^{\infty} d\tilde{\phi}\, e^{-\beta R^d \epsilon^{2-d/2}(\pm\tilde{\phi}^2+\tilde{\phi}^4)}. \tag{88}$$

For $G = R^d\epsilon^{2-d/2} \gg 1$ we can do the integral in Eq. (88) using saddle point techniques. For $\epsilon < 0$ the saddle points are at $\phi = \pm 1/\sqrt{2}$, and we obtain

$$Z(\epsilon) \propto e^{\beta R^d \epsilon^{2-d/2}}, \tag{89}$$

and hence the free energy is

$$-k_B T \ln Z(\epsilon) = -R^d\epsilon^{2-d/2}. \qquad (G \gg 1) \tag{90}$$

We have neglected the logarithmic corrections generated by the steepest descent integral. Note the minus sign on the right-hand side of Eq. (90). For $\epsilon > 0$ the saddle points are at $\pm i/\sqrt{2}$ and the free energy is also proportional to $-R^d\epsilon^{2-d/2}$. Hence as argued in Sec. V and Appendix A we can use the percolation mapping to show that the number of fundamental clusters scales as $R^d\epsilon^{2-d/2}$ near the critical point. As usual, similar arguments can be used near the spinodal for $G_s \to \infty$ and near the pseudospinodal for $G_s \gg 1$.

To determine the lifetime of the fundamental clusters, we return to the Parisi-Sourlas method. At the spinodal we have from Eq. (67)

$$\int d\mathbf{x}dt \Big(\frac{\partial\psi}{\partial t}\Big)^2 \sim 1. \tag{91}$$

As in Sec. III the lifetime of a fundamental cluster is found by requiring that

$$\frac{\psi_{\rm fc,s}^2\,\xi^d}{\tau_{\rm fc,s}} \sim \frac{(\Delta h^{1/2})^2 R^d \Delta h^{d/4}}{[R^d\Delta h^{3/2-d/4}]^2\,\tau_{\rm fc,s}} \sim 1, \tag{92}$$

or

$$\tau_{\rm fc,s} \sim \frac{\Delta h^{-1/2}}{R^d\Delta h^{3/2-d/4}}. \quad \text{(fundamental cluster lifetime near spinodal)} \tag{93}$$

We see that near the critical point ($G \gg 1$) and the pseudospinodal ($G_s \gg 1$), the lifetime of the fundamental clusters is considerably shorter than the lifetime (decorrelation time) of a fluctuation near the critical point (ϵ^{-1}) and the spinodal ($\Delta h^{-1/2}$).

To understand the relation between the lifetime of the fundamental clusters and the lifetime of a critical phenomena fluctuation recall that the clusters are independent. We consider the fluctuations to be formed from the "vacuum" (zero magnetization near the

critical point and zero net magnetization after the infinite cluster is subtracted near the spinodal) by a random walk in the number of fundamental clusters. At the critical point the density of the critical phenomena fluctuations is given by Eq. (17). Because the fluctuations arise from a random walk in the number of fundamental clusters, there must be a "walk" of $(R^d\epsilon^{2-d/2})^{1/2}$ cluster flips (steps) in the direction of the fluctuation to obtain a density of

$$\phi_f(\mathbf{x}) \sim \frac{\epsilon^{1/2}}{R^d\epsilon^{2-d/2}}(R^d\epsilon^{2-d/2})^{1/2} = \frac{\epsilon^{1/2}}{(R^d\epsilon^{2-d/2})^{1/2}}. \tag{94}$$

The time needed for $R^d\epsilon^{2-d/2}$ attempted cluster flips is

$$\tau_{\rm f,c} \sim \frac{\epsilon^{-1}}{R^d\epsilon^{2-d/2}} R^d\epsilon^{2-d/2} = \epsilon^{-1}, \tag{95}$$

in agreement with Eq. (37). The same considerations near the spinodal yield $\tau_{\rm f,s} \sim \Delta h^{-1/2}$ for the lifetime of a fluctuation, in agreement with our earlier result for $\tau_{\rm f,s}$ in Eq. (68).

In summary, we have argued that the fluctuations near a mean-field critical point and a spinodal are not represented by a single fundamental cluster. The relation between these clusters and the fluctuations in (near-)mean-field systems is qualitatively different than in systems that obey hyperscaling. In the former the fluctuations are formed by a random walk in the number of fundamental clusters that "flip" on a time scale much shorter than the scale set by critical slowing down. A summary of our notation and our main results so far is given in Table I.

VII. CLUSTER STRUCTURE AND INSTABILITIES IN SUPERCOOLED LIQUIDS

We begin our discussion of the consequences of the fluctuation structure in (near-)mean-field systems by considering the liquid-solid spinodal in supercooled fluids. To explain the role of the structure of the fluctuations we first provide some background. In 1951 Kirkwood [42] noted that approximate equations for the distribution functions in the liquid state appeared to show an instability as the supercooled liquid is quenched deeper. Kirkwood began with the first equation of the static BBGKY hierarchy [43]

$$-k_BT\nabla_1\rho^{(1)}(\mathbf{x}_1) = \int d\mathbf{x}_2\, \nabla_1 V(x_{12})\rho^{(2)}(\mathbf{x}_1,\mathbf{x}_2), \tag{96}$$

where $\rho^{(1)}(\mathbf{x}_1)$ and $\rho^{(2)}(\mathbf{x}_1,\mathbf{x}_2)$ are the one and two particle distribution functions respectively, ∇_1 denotes differentiation with respect to the position of particle 1, and the interaction

quantity	mean-field critical point	spinodal	hyperscaling
Ginzburg parameter	$G = R^d \epsilon^{2-d/2}$	$G_s = R^d \Delta h^{3/2-d/4}$	$G\,(G_s)$ fixed
order parameter	$\phi \sim \epsilon^{1/2}$	$\psi \sim \Delta h^{1/2}$	$\sim \epsilon^{\beta}$
fluctuations in order parameter density	$\phi_{\rm f}(\mathbf{x} \lesssim \xi) \sim \epsilon^{1/2}/G^{1/2}$	$\psi_f(\mathbf{x} \lesssim \xi) \sim \epsilon^{1/2}/G_s^{1/2}$	$\sim \epsilon^{\beta}$
lifetime of fluctuation	$\tau_{\rm f,c} \sim \epsilon^{-1}$	$\tau_{\rm f,s} \sim \varepsilon^{-1}$	$\sim \epsilon^{-z/\nu}$
lifetime of fundamental cluster	$\tau_{\rm fc,c} \sim \epsilon^{-1}/G$	$\tau_{\rm fc,s} \sim \varepsilon^{-1}/G_s$	
mean number of clusters in correlation length volume	$\overline{n}_{\rm fc,c} \sim G$	$\overline{n}_{\rm fc,s} \sim G_s$	~ 1
density of fundamental cluster	$\phi_{\rm fc,c}(\mathbf{x} \lesssim \xi) \sim \epsilon^{1/2}/G$	$\psi_{\rm fc,s}(\mathbf{x} \lesssim \xi) \sim \epsilon^{1/2}/G_s$	
mean number of excess fundamental clusters	$\overline{\Delta}_{\rm fc,c} \sim G^{1/2}$	$\overline{\Delta}_{\rm fc,s} \sim G_s^{1/2}$	

TABLE I: Summary of our notation and some of the important scaling relations derived in the text. The spinodal can be approached by reducing the magnetic field difference Δh for fixed temperature or by decreasing the temperature difference ε for fixed magnetic field. The two approaches are related by $\Delta h^{1/2} \sim \varepsilon$. The exponent z characterizes critical slowing down and is of order 2 for systems described by model A [34].

potential $V(x_{12})$ is assumed to be pairwise additive and spherically symmetric. Suppose that the system is in the liquid phase where $\rho^{(1)}(\mathbf{x}_1)$ is a constant equal to ρ, and $\rho^{(2)}(\mathbf{x}_1, \mathbf{x}_2)$ is a function of $x_{12} = |\mathbf{x}_1 - \mathbf{x}_2|$ and is equal to $\rho^2 h(x_{12}) = \rho^2(1 + g(x_{12}))$, where $g(x_{12})$ is the pair correlation function [43]. We substitute

$$\rho^{(1)}(\mathbf{x}_1) = \rho + \omega(\mathbf{x}_1) \tag{97}$$

into Eq. (96), treat $\omega(\mathbf{x}_1)$ as a small perturbation, and linearize Eq. (96) to find

$$-k_B T \frac{\nabla_1 \omega(\mathbf{x}_1)}{\rho} = \int d\mathbf{x}_2 \, \nabla_1 V(x_{12}) h(x_{12}) \omega(\mathbf{x}_2), \tag{98}$$

where the spherical symmetry of $V(x_{12})$ and $h(x_{12})$ results in

$$\int d\mathbf{x}_2 \, \nabla_1 V(x_{12}) h(x_{12}) = 0. \tag{99}$$

We have also assumed that a possible instability in $h(x_{12})$ is higher order in ω. If we define

$$q(\mathbf{x}_1 - \mathbf{x}_2) = \nabla_1 V(x_{12}) h(x_{12}), \tag{100}$$

we see that there is an instability [42] if there is a nonzero solution to

$$k_B T \nabla_1 \omega(\mathbf{x}_1) + \beta\rho \int d\mathbf{x}_2 \, q(\mathbf{x}_1 - \mathbf{x}_2)\omega(\mathbf{x}_2) = 0. \tag{101}$$

Kirkwood analyzed Eq. (101) for the hard sphere fluid and found that there was an instability in $d = 3$ [42]. However, he ignored a possible instability in $h(x_{12})$ which is related by the BBGKY [43] hierarchy to possible instabilities in all of the distribution functions. Hence, it is not clear that the instability Kirkwood found is real. A more careful analysis [44] suggests that the instability vanishes when higher order terms are considered.

To investigate the existence of an instability and its relation to a possible spinodal, Grewe and Klein [45, 46] investigated the properties of a simple fluid for which the interaction potential has the Kac form [5] given in Eq. (1) with $V_R = 0$ and

$$\Phi(\gamma|\mathbf{x}|) = \begin{cases} 1 & \text{if } \gamma|\mathbf{x}| \le 1, \\ 0 & \text{if } \gamma|\mathbf{x}| > 1. \end{cases} \tag{102}$$

In the mean-field limit $\gamma \to 0$, it was shown that all distribution functions of order higher than two are completely specified by only the single particle and pair distribution functions and that $\rho^{(1)}(\mathbf{x}_1)$ in the limit $\gamma \to 0$ satisfies the equation [45, 46]

$$\rho^{(1)}(\mathbf{x}_1) = z \exp[-\beta \int d\mathbf{x}_2 \, \Phi(|\mathbf{x}_{12}|)\rho^{(1)}(\mathbf{x}_2)], \tag{103}$$

where $z = e^{-\beta\mu}$ and μ is the chemical potential. Similarly, $g(|\mathbf{x}_{12}|)$ satisfies

$$g(|\mathbf{x}_{12}|) = \beta\rho\Phi(|\mathbf{x}_{12}|) - \beta\rho \int d\mathbf{x}_3 \, g(|\mathbf{x}_1 - \mathbf{x}_3|)\Phi(|\mathbf{x}_2 - \mathbf{x}_3|), \tag{104}$$

where all length scales are in units of $\gamma^{-1} = R$. Note that $g(|\mathbf{x}_{12}|)$ is of order γ^d. The derivations of Eqs. (103) and (104) are given in Refs. 45 and 46.

From Eq. (104) the structure function $S(k)$, which is obtained by taking the Fourier transform of $g(|\mathbf{x}_{12}|)$, is proportional to

$$S(k) \propto \frac{1}{1 + \beta\rho\hat{\Phi}(k)}, \tag{105}$$

where $\hat{\Phi}(k)$ is the Fourier transform of $\Phi(\gamma|\mathbf{x}|)$ and $k = |\mathbf{k}|$. Note that the structure function is order one in the $\gamma \to 0$ limit.

We can perform a stability analysis on Eq. (103) similar to that done by Kirkwood on Eq. (96). We substitute Eq. (97) into Eq. (103) and linearize in $\omega(\mathbf{x}_1)$ to obtain

$$\omega(\mathbf{x}_1) = -\beta\rho \int d\mathbf{x}_2 \, \Phi(|\mathbf{x}_1 - \mathbf{x}_2|)\omega(\mathbf{x}_2), \tag{106}$$

where ρ is the solution of $\rho = z \exp[-\beta\hat{\Phi}(0)\rho]$, and $\hat{\Phi}(0) = \int d\mathbf{x} \, \Phi(|\mathbf{x}|) > 0$.

There is an instability only if there is a nonzero solution to Eq. (106). If we take the Fourier transform of Eq. (106), we can express the instability condition as

$$1 + \beta\rho\hat{\Phi}(|\mathbf{k}|) \leq 0, \tag{107}$$

or $\hat{\Phi}(|\mathbf{k}|) < 0$ for some value of $|\mathbf{k}|$. This condition is satisfied for the potential in Eq. (102). Because $\Phi(\gamma|\mathbf{x}|)$ in Eq. (102) has a Fourier transform that is bounded from below, there is a value of $\beta\rho$ below which there is no instability. If $k_0 = |\mathbf{k}|$ is the location of the global minimum of $\hat{\Phi}(|\mathbf{k}|) < 0$, then the system has no instability for $\beta\rho < -1/\hat{\Phi}(k_0)$. We see from Eq. (105) that the structure function $S(k_0)$ first diverges for fixed ρ (as T is decreased) at the same value of the temperature T at which an instability first appears. The divergence of $S(k_0)$ implies that the instability in the mean-field system is a spinodal and is analogous to the divergence of the susceptibility at the Ising spinodal.

In the mean-field limit no higher order distribution functions need to be considered and the results of Grewe and Klein [45, 46] are rigorous. The structure function $S(k_0)$ diverges as $(T - T_s)^{-1}$ so that the critical exponent is the same as the Ising spinodal if the temperature rather than the magnetic field is used to approach the spinodal in the Ising model. The only difference is that $S(k)$ diverges at $k = 0$ in Ising models and gases rather than at $k_0 \neq 0$. The other critical exponents are also the same as for the Ising spinodal [45, 46].

We next discuss an important difference between measurements of the spinodal exponents in Ising models and in supercooled fluids. As we have discussed (see Fig. 1), there is no spinodal in an Ising model for R finite, but we see spinodal-like behavior for $R \gg 1$ if the system is not quenched too deeply into the metastable state [7, 8, 9]. The larger R, the more the pseudospinodal behaves like a true spinodal.

In the supercooled liquid there is no direct evidence of a spinodal or pseudospinodal from either experiments or simulations. We will see that this lack of direct evidence is due to

the structure of the fluctuations in (near-)mean-field systems and the crucial role of the fundamental clusters.

For the potential in Eq. (102) the system is a fluid for high temperatures and/or low densities [45, 46, 47]. If the temperature T is lowered at a fixed density, the liquid-solid instability is encountered at the spinodal temperature T_s. If T is lowered below T_s, the uniform density fluid phase becomes unstable and a "clump" phase is formed [47]. Because we are interested in the nature of the spinodal, the behavior of the system for $T < T_s$ is not of interest here.

Unlike Ising/Potts models, there is no precise definition of a cluster in a continuum system of particle. However, it is reasonable to assume that the scaling behavior of the fluctuations and fundamental clusters near the liquid-solid spinodal is the same as near the Ising spinodal. This assumption is consistent with the fact that the Ising and spinodal exponents for the system defined by Eq. (102) are the same. For convenience, we will use temperature scaling near the liquid-solid transition rather than the analog of magnetic field scaling. From Eqs. (93) and (64) the lifetime of the fundamental clusters as $\varepsilon \to 0$ scales as

$$\tau_{\rm fc,s} \sim \frac{\varepsilon^{-1}}{R^d \varepsilon^{3-d/2}}. \qquad \text{(lifetime of fundamental cluster near spinodal)} \tag{108}$$

Grewe and Klein [45, 46] used the mean-field formalism developed by Kac et al. [5]. In particular, the interaction range $R = \gamma^{-1}$ is taken to infinity before the spinodal is approached, that is, before the limit $\varepsilon \propto (T - T_s)$ goes to zero. Hence, in the mean-field limit there exists fundamental clusters with probability of order one but zero lifetime (see the discussion in Sec. III). In experiments and simulations, the converse is true. For example, consider a measurement of the structure function $S(k)$ in a simulation. The structure function is obtained by computing

$$S(k) = \frac{1}{N} \langle \big[\sum_j e^{i\mathbf{k}\cdot\mathbf{x}_j} \big]^2 \rangle, \tag{109}$$

where $\mathbf{x}_j$ is the instantaneous position of particle j and $\langle \cdots \rangle$ denotes an ensemble average. Because a simulation can be performed only on systems with finite R, the time of the measurement, which is instantaneous, is much less than the nonzero fundamental cluster lifetime in Eq. (108). Therefore the result of the simulations need not be consistent with the mean-field predictions [45, 46]. (The time scale of the measurement does not refer to

the time over which data is taken, but to the time during which the probe (say a neutron) is in contact with a fluctuation.)

Before we consider the behavior of $S(k_0)$ in supercooled liquids, we discuss the measurement of $S(k)$ near the Ising spinodal. We will find that the measured behavior of $S(k=0)$ agrees with the mean-field predictions [35]. The application of the scaling argument in Sec. V to the susceptibility near the Ising spinodal gives (see Eqs. (18) and (81))

$$\chi \sim \psi_f^2 \, \xi^d \sim \Big[\frac{\Delta h^{1/2}}{(R^d \Delta h^{3/2-d/4})^{1/2}}\Big]^2 R^d \Delta h^{-d/4} \sim \Delta h^{-1/2}. \tag{110}$$

Alternatively, we can calculate χ directly from the fundamental clusters. The density of a fundamental cluster is given in Eq. (80). Hence, the isothermal susceptibility associated with one fundamental cluster is (see Eq. (81))

$$\chi_{1\rm fc} \sim \psi_{fc,s}^2 \, \xi^d \sim \Big[\frac{\Delta h^{1/2}}{R^d \Delta h^{3/2-d/4}}\Big]^2 R^d \Delta h^{-d/4} \sim \frac{\Delta h^{-1/2}}{R^d \Delta h^{3/2-d/4}}. \tag{111}$$

Because the clusters are independent, the isothermal susceptibility of the system is the sum of the individual cluster isothermal susceptibilities. Because there are $R^d \Delta h^{3/2-d/4}$ clusters in a correlation length volume, $\chi = \Delta h^{-1/2}$, consistent with Eq. (110). Although there are clusters of both up and down spins, each cluster has the same isothermal susceptibility because the susceptibility is proportional to the square of the spin density [32, 33].

Another way of obtaining the same result as in Eq. (110) is to calculate the structure function $S_{1\rm fc}(\mathbf{k})$ for one fundamental cluster from the Fourier transform of the connectedness function of the cluster. We can write (see Eq. (80))

$$S_{1{\rm fc,\,s}}(\mathbf{k}) \sim \Big[\frac{\Delta h^{1/2}}{R^d \Delta h^{3/2-d/4}}\Big]^2 \delta(\mathbf{k}). \tag{112}$$

The delta function comes from integrating over the infinite size of the spatially uniform cluster. For a cluster with the spatial extent of the correlation length ξ, $\delta(\mathbf{k})$ in Eq. (112) would be replaced by a function whose height is ξ^d and whose width is proportional to ξ^{-1}.

The difference between the fundamental cluster lifetime $\tau_{\rm fc,s}$ in Eq. (93) and the lifetime of a fluctuation $\sim \Delta h^{-1/2}$ suggest the following picture. A fluctuation is a collection of fundamental clusters that appear and disappear on the time scale $\tau_{\rm fc,s}$. The lifetime of a fluctuation is a much longer than a fundamental cluster for $G_s \gg 1$, which implies that the fundamental clusters come and go with different angular orientations but with their centers in roughly the same place for a time of order $\Delta h^{-1/2}$, the lifetime of a fluctuation. If we

make a measurement on a time scale $\gg \Delta h^{-1/2}$, the number of up and down fundamental clusters would be the same on the average, and the measured structure function would show no correlations between the spins.

Consider a measurement on a time scale $t_{\rm meas}$ such that $t_{\rm meas} \gg \tau_{\rm fc,s}$ and $t_{\rm meas} \sim \Delta h^{-1/2}$. Because the measurement time is comparable to the lifetime of the fluctuation, the measurement will see a spin density equal to the fluctuation density, which is generated by fluctuations in the number of fundamental clusters. That is, the individual fundamental clusters cannot be distinguished, and

$$S(\mathbf{k}) \sim \Big[\frac{\Delta h^{1/2}}{(R^d \Delta h^{3/2-d/4})^{1/2}}\Big]^2 \delta(\mathbf{k}). \tag{113}$$

If we replace $\delta(\mathbf{k})$ by ξ^d, we obtain the same result as in Eq. (110), obtained by considering the fluctuations directly.

Now suppose that a measurement is made such that $t_{\rm meas} \ll \tau_{\rm fc,s}$. In this case an external probe or a simulation would see a set of order $R^d \Delta h^{3/2-d/4}$ *frozen* fundamental clusters. To determine the structure function that would be measured, we need to add the cluster structure function in Eq. (112) for the $R^d \Delta h^{3/2-d/4}$ frozen clusters. To do so we convert the sum to an integral by using one of the factors of $1/R^d \Delta h^{3/2-d/4}$ in Eq. (112) to create an infinitesimal element of solid angle $d\Omega$. (We ignore numerical factors because we are interested only in the scaling properties.) In so doing we are assuming that the $R^d \Delta h^{3/2-d/4}$ fundamental clusters overlap each other with random orientations. Hence the sum over clusters becomes an integral over solid angle:

$$S(|\mathbf{k}|) \sim \int d\Omega \, \frac{\Delta h}{R^d \Delta h^{3/2-d/4}} \delta(\mathbf{k}). \tag{114}$$

For $d = 3$ we have in spherical polar coordinates

$$S(k) \sim \frac{\Delta h}{R^3 \Delta h^{3/2-3/4}} \int \sin\theta d\theta d\phi \, \frac{\delta(k)\delta(\theta)\delta(\phi)}{k^2 \sin\theta}. \tag{115}$$

Hence

$$S(k) \sim \frac{\Delta h}{R^3 \Delta h^{3/2-3/4}} \frac{\delta(k)}{k^2}. \tag{116}$$

If we now replace $\delta(k)$ by ξ with $k \sim \xi^{-1}$, we obtain

$$S(k) \sim \frac{\Delta h}{R^3 \Delta h^{3/2-3/4}} \xi^3 = \frac{\Delta h}{R^3 \Delta h^{3/2-3/4}} R^3 \Delta h^{-3/4} = \Delta h^{-1/2}, \tag{117}$$

in agreement with Eq. (110). The generalization to arbitrary dimensions is straightforward. We conclude that we obtain the same scaling dependence for the susceptibility (equal to $S(k = 0)$), independent of the relative order of magnitude of the measurement time and consistent with simulations of the Ising model [7].

We now discuss the measurement of $S(k)$ near the liquid-solid spinodal. As stated, we assume that the fundamental clusters in the supercooled liquid scale the same way as they do in Ising models. Because the clusters are independent [38], the structure function of a single fundamental cluster has to contain information about the symmetry of the instability. That is, a collection of independent clusters cannot generate a symmetry that does not already exist in each cluster. Because we are interested in the limit of stability of the supercooled liquid and know that the instability occurs at $k_0 \neq 0$, the clusters must reflect this symmetry. Hence, we will assume that the clusters have a symmetry reflected by the wave vector $\mathbf{k}_0$ with arbitrary orientation. In analogy with the Ising spinodal, we expect that the structure function $S_{\rm fc,s}(\mathbf{k})$ of the fundamental clusters can be approximated near the spinodal by

$$S_{\rm fc,\,s}(\mathbf{k}) \sim \Big[\frac{\varepsilon}{R^d\varepsilon^{3-d/2}}\Big]^2 \delta(\mathbf{k}-\mathbf{k}_0). \tag{118}$$

We have used temperature variables rather than the chemical potential, the analog of the magnetic field. There are other peaks in $S_{\rm fc,\,s}(\mathbf{k})$ at $|\mathbf{k}| \neq \mathbf{k}_0$, but we will focus on $S_{\rm fc,\,s}(\mathbf{k}_0)$, the peak associated with the divergence as the liquid-solid spinodal is approached.

We first consider a measurement on a time scale such that $t_{\rm meas} \ll \tau_{\rm fc,s}$, where $\tau_{\rm fc,s}$ is give by Eq. (108). As before we need to sum over all orientations of the frozen clusters whose centers are fixed. We convert this sum to an integral by absorbing one of the factors of $1/R^d\varepsilon^{3-d/4}$ in Eq. (118) to form an infinitesimal. In this case we need to integrate over orientations of the vector $\mathbf{k}_0$ keeping the magnitude k_0 constant. For $d = 3$ we have

$$S(k) \sim \frac{\varepsilon^2}{R^3\varepsilon^{3-3/2}}\frac{1}{k_0^2}\iint d\theta d\phi\, \delta(k-k_0)\delta(\theta)\delta(\phi). \tag{119}$$

The factor of $\sin\theta$ in the numerator associated with the solid angle and the $\sin\theta$ in the denominator associated with the delta function in spherical polar coordinates cancel. The integrals in Eq. (119) give

$$S(k) \sim \frac{\varepsilon^2}{R^3\varepsilon^{3-3/2}}\frac{1}{k_0^2}\delta(k-k_0). \tag{120}$$

As for the Ising case we replace the delta function by the correlation length $\xi \sim R\varepsilon^{-1/2}$.

Hence, the structure function at $k = k_0$ scales as

$$S(k_0) \sim \frac{\varepsilon^2 R \varepsilon^{-1/2}}{R^3 \varepsilon^{3-3/2}} = R^{-2}\varepsilon^0. \qquad (d = 3) \tag{121}$$

Equation (121) implies that there is either no divergence or the divergence is logarithmic, which is consistent with the fact that no direct evidence of a pseudospinodal has been observed in simulations of simple fluids in $d = 3$, despite the indirect evidence that nucleation is influenced by a pseudospinodal in deeply quenched Lennard-Jones liquids [16, 48, 49] and in nickel [17].

It is easy to show that $S(k_0)$ for arbitrary dimensions scales as

$$S(k_0) \sim \frac{\varepsilon^2 R \varepsilon^{-1/2}}{R^d \varepsilon^{3-d/2}} = R^{-d+1}\varepsilon^{-3/2+d/2} \propto \varepsilon^{-\tilde{\gamma}}. \tag{122}$$

Equation (122) predicts that $\tilde{\gamma} = 1$ for $d = 1$, $\tilde{\gamma} = 1/2$ for $d = 2$, and $\tilde{\gamma} = 0$ for $d = 3$, in contrast to the mean-field result $\gamma = 1$ for all d (see Eq. (105)). Results consistent with the predictions in Eq. (122) were found in $d = 1$–3 for the potential in Eq. (102) [50].

If we could do a measurement on a time scale of the order of the fluctuation lifetime ε^{-1}, we would see a smeared out density fluctuation that was radially symmetric and varied periodically in the radial direction. We expect that the dominant periodicity would be characterized by the wave vector k_0 and that the divergent contribution to the structure function near the spinodal could be approximated for $k \approx k_0$ by

$$S_{\mathrm{f,s}}(k) \sim \frac{\varepsilon^2}{R^d \varepsilon^{3-d/2}} \int dx\, x^{d-1} e^{i(k-k_0)x}. \tag{123}$$

Note that we used the density of a fluctuation rather than the cluster, which is appropriate for the time averaged cluster distribution. At $k = k_0$ the structure function will be given by

$$S_{\mathrm{f,s}}(k_0) \sim \frac{\varepsilon^2}{R^d \varepsilon^{3-d/2}} R^d \varepsilon^{-d/2} = \varepsilon^{-1}, \tag{124}$$

consistent with the results of Refs. 45 and 46.

In summary, the structure and finite lifetime of the fundamental clusters are responsible for the behavior of $S(k)$ near the liquid-solid spinodal. If the peak of $S(k)$ is at $k = 0$ as in the Ising model, the fact that measurements are made on a time scale short compared to the fundamental cluster lifetime rather than a time scale much longer as required by mean-field theory makes no difference to the measured value of the exponent that characterizes the divergence. In contrast, if the peak of $S(k)$ is at $k \neq 0$, the measurement time scale affects the observed value of the exponent.

VIII. CLUSTER STRUCTURE AND NUCLEATION

In classical nucleation theory [19] the nucleating droplet is assumed to be isolated, compact, and describable as a fluctuation about a quasiequilibrium metastable state. Classical nucleation occurs near the coexistence curve independent of the range of interaction [51]. For systems with sufficiently long-range interactions a quench near the pseudospinodal can lead to nucleation being influenced by the critical point nature of the pseudospinodal, which implies that the surface tension will vanish as the spinodal is approached. Near the pseudospinodal the surface tension will be nonzero, nucleation will occur with a very small surface tension [11], and the nucleating droplet is no longer compact as in classical nucleation theory [10, 11]. We refer to this form of nucleation as *spinodal nucleation*.

An elegant way to approach nucleation theoretically was developed most fully by Langer [18, 19] and adapted to spinodal nucleation by Klein and Unger [11, 52]. The Hamiltonian in Eq. (4) is used to calculate the free energy $F(\beta, h)$ in the equilibrium state

$$F(\beta, h) = -k_B T \ln \int \delta\phi \, e^{-\beta H(\phi)}, \tag{125}$$

which can be analytically continued from the stable to the metastable state [18]. The nucleating droplet is associated with the solution of the Euler-Lagrange equation obtained from the functional derivative of the Hamiltonian [18]:

$$-R^2 \nabla^2 \phi(\mathbf{x}) - 2|\epsilon|\phi(\mathbf{x}) + 4\phi^3(\mathbf{x}) - h = 0. \tag{126}$$

In Eq. (126) $\epsilon < 0$, that is, the temperature T is below the critical temperature. The dominant exponential part of the nucleation probability is obtained by substituting the solution to Eq. (126) into the expression for the imaginary part of the analytically continued free energy. The details of this approach can be found in Ref. 18 and are outlined in Ref. 19.

Klein and Unger expanded the Hamiltonian in Eq. (4) about the mean-field spinodal as in Eq. (52) and obtained an Euler-Lagrange equation of the form [52]

$$-R^2 \nabla^2 \psi(\mathbf{x}) + \lambda_1 \Delta h^{1/2} \psi(\mathbf{x}) - \lambda_2 \psi^2(\mathbf{x}) = 0, \tag{127}$$

where λ_1 and λ_2 are constants for fixed ϵ. It is straightforward to show that the solution to Eq. (127) must have the form [11, 52]

$$\psi(\mathbf{x}) = \Delta h^{1/2} f\Big(\frac{\mathbf{x}}{R \Delta h^{-1/4}}\Big), \tag{128}$$

which implies that the difference in the order parameter density between the interior of the droplet and the background is $\sim \Delta h^{1/2}$; this difference is vanishingly small for large R and small Δh. This small difference presents two problems: how can we identify when and where the nucleating droplet occurs and how can we determine its structure. In Ising models these problems have been solved by mapping the spinodal onto a percolation transition and using the relation between the clusters and the nucleating droplet.

Because the density of the nucleating droplet over the background is the order of $\Delta h^{1/2}$, the density of the fundamental clusters is order $\Delta h^{1/2}/R^d \Delta h^{3/2-d/4}$ (Eq. (80)), and the density of a fluctuation is $\Delta h^{1/2}/(R^d \Delta h^{3/2-d/4})^{1/2}$, the nucleating droplet is not a fundamental cluster or a fluctuation described by the Gaussian approximation. From the discussion in Sec. VI we know that the fluctuations near a spinodal are generated by a random walk of the number of fundamental clusters in the up or down direction. A possible scenario might be that the nucleating droplet is generated by a random walk in the number of fundamental clusters that produces a region of density $\Delta h^{1/2}$. From the discussion in Secs. V and VI such a process would require a "walk" of distance $R^d \Delta h^{3/2-d/4}$ ($[R^d \Delta h^{3/2-d/4}]^2$ steps) and a time of $\tau \sim \Delta h^{-1/2} R^d \Delta h^{3/2-d/4}$. Because the nucleation time is the inverse of the probability [18], the nucleation time would be of the order of $\exp(\beta R^d \Delta h^{3/2-d/4})$. Hence a random walk occurs too quickly to account for the time needed to see nucleation.

A clue to the relation between the fundamental clusters and the nucleating droplets is provided in Ref. 12 in which nucleation was observed near the spinodal in a $d = 2$ Ising model with long-range interactions. The nucleating droplet was identified using intervention, and it was found that the number of fundamental clusters in a correlation length volume just prior to nucleation is the order of G_s, implying that G_s fundamental clusters with density $\Delta h^{1/2}/G_s^{1/2}$ coalesced into an object with a density on the order of $\Delta h^{1/2}$.

The results of Ref. 12 together with the random walk argument suggests the following picture of the relation between the fundamental clusters and the nucleating droplets. While the system is in the metastable state, there are fluctuations in the number of fundamental clusters. These fluctuations result in regions with order $G_s = R^d \Delta h^{3/2-d/4}$ fundamental clusters in excess of the background on a time scale $\sim R^d \Delta h^{3/2-d/4} \Delta h^{-1/2}$. Because this time scale is much less than the time scale for nucleation, which is of the order of $\exp(\beta R^d \Delta h^{3/2-d/4})$ [11], the appearance of G_s clusters with a linear spatial extent of the correlation length will happen everywhere in the system many times before the nucleation event. In Fig. 3 we plot

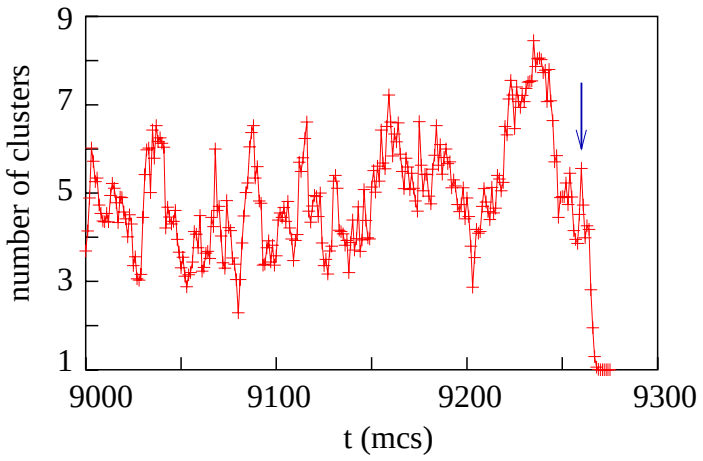

FIG. 3: The number of fundamental clusters in a region the size of the correlation length where nucleation will occur (see arrow). Note the number of times where the number of fundamental clusters is of order G_s. The Monte Carlo simulation was done for a $d = 2$ Ising model with $R = 28$, $h = 1.25$, $G_s = 12.3$, and linear dimension $L = 560$.

the number of fundamental clusters the size of the correlation length in a region where we know that nucleation will occur. Notice the number of time intervals where the number of fundamental clusters is of order G_s.

Because the time scale for nucleation is of the order of $\exp(\beta R^d \Delta h^{3/2-d/4})$ and G_s fundamental clusters coalesce at nucleation, there must be a free energy cost associated with the coalescence that is the order of G_s. We can estimate this free energy cost by noting that the coalescence of G_s fundamental clusters change the density only infinitesimally. This assumption is justified because the random walk is assumed to generate a density in the stable phase direction of order $\Delta h^{1/2}$ and the nucleating droplet has the same density. Therefore the energy change due to coalescence is negligible. The entropy cost can be estimated by noting that there are G_s fundamental clusters before nucleation and one nucleating droplet after coalescence. These considerations imply that the entropy change is given by

$$\Delta S \sim \ln 2 - \ln 2^{R^d \Delta h^{3/2-d/4}} \sim -R^d \Delta h^{3/2-d/4} \ln 2, \tag{129}$$

because each cluster has two states, up and down. Because the energy change is negligible, the free energy change due to coalescence is the order of $G_s \gg 1$. Hence the probability of nucleation and hence the probability of coalescence is $\propto \exp(-\beta \Delta F) \sim \exp(-\beta G_s)$, in agreement with Refs. 11 and 52.

This discussion and the one in Sec. VII suggests that the fundamental clusters are not only a mathematical construct, but are real physical objects whose probability is not given by the usual Boltzmann factor (see Sec. III). This suggestion will be given further credence by the discussion in the next section.

IX. CLUSTERS AND MODELS OF EARTHQUAKE FAULTS

In this section we discuss the relevance of the cluster structure to our understanding of models of earthquake faults. The original Burridge-Knopoff model consists of blocks connected by linear springs to their nearest neighbors with spring constant k_c [53]. The blocks are also connected to a loader plate by linear springs with spring constant k_L, and rest on a surface with a nonlinear velocity-weakening stick-slip friction force.

A simulation is initiated by choosing the displacements of the blocks at random. While the loader plate is fixed, we determine the stress on each block (the force due to the springs) and update its velocity and displacement according to Newton's equations of motion. We continue these updates until all blocks are stuck. A block is "stuck" when its velocity is below a certain threshold and other criterion are met [54, 55, 56]. We then add stress to all the blocks by moving the loader plate to bring the block with the largest stress to failure. That is, when the stress on a block exceeds the static coefficient of friction, the block "fails" and begins to slip. This process insures that there is only one block that initiates the failure sequence. An earthquake is comprised of all the blocks that fail between plate updates.

The number n_s of earthquakes with s blocks exhibits a power law dependence on s,

$$n_s \sim s^{-x}, \tag{130}$$

with $x \approx 2$ if the blocks are connected by only nearest-neighbor springs [54, 55, 56]. However, the calculation of realistic stress transfer Green's functions for real faults [57, 58] suggests that we should consider springs that connect further neighbor blocks. The behavior of the generalized Burridge-Knopoff model is more complicated and in the limit of long-range stress transfer and a slow decrease of the velocity-weakening friction force with increasing velocity, it has been found in simulations that $x \approx 3/2$ [56].

To provide more insight into the behavior of the model with long-range stress transfer we discuss a cellular automaton (CA) version of the Burridge-Knopoff model introduced

by Rundle, Jackson, and Brown [59, 60, 61], which considers blocks and springs as in the Burridge-Knopoff model. A failure threshold σ_F and a residual stress σ_R is specified for each block. For simplicity, we will take the σ_F and σ_R to be the same for all blocks. The stress on a block is given by $\sigma_j = k_L(\Delta - u_j) + k_c \sum_i (u_i - u_j)$, where Δ is the displacement of the loader plate and u_j is the displacement of block j from its initial position. If $\sigma_j < \sigma_F$, we do nothing and proceed to the next block. If $\sigma_j \geq \sigma_F$, we move the block a distance Δu where

$$\Delta u_j = \frac{\sigma_j - \sigma_R}{k_L + q k_c}, \tag{131}$$

and q is the number of sites within the interaction range. If the range of stress transfer is R, then $q = (2R+1)^2$. Once the system is quiescent, that is, $\sigma_j < \sigma_F$ for all j, the plate is updated as in the Burridge-Knopoff model.

The number of earthquakes with s failed blocks has a power law dependence as in Eq. (130) with $x = 3/2$ in the mean-field limit $R \to \infty$. How does this scaling arise? A clue is that the long-range stress transfer CA models with noise added to the stress drop can be described by equilibrium statistical mechanics [62, 63]. It also has been shown that this model can be described by a Langevin or Landau-Ginzburg equation in the limit $R \to \infty$ [64]. The latter equation has the same form as Eq. (52) (see Ref. 3). These considerations imply that the scaling behavior $n_s \sim s^{-3/2}$ in the long-range CA models is identical for scaling purposes to scaling near the Ising spinodal [3].

To obtain the scaling exponent x, we use the fact that the Ising spinodal can be described by a Fisher droplet model [65] in which the system near a critical point can be described by a collection of non-interacting droplets. Fisher assumed that the distribution of the droplets scale as [65]

$$\tilde{n}_s \sim \frac{e^{-\Delta h s^{\tilde{\sigma}}}}{s^{\tau}}. \tag{132}$$

From the cluster mapping near the spinodal discussed in Appendix A, we know that there exists independent objects, the fundamental clusters, which scale in a similar way. To calculate the exponent τ for the earthquake CA model, we need only calculate the cluster scaling exponent for the Ising spinodal.

To obtain the exponent τ, which we will relate to the exponent x in Eq. (130), we note that the Fisher droplet model exponents are related to the spinodal exponents through the first several moments of Eq. (132). In particular, the isothermal susceptibility χ is given by

the second moment of $\tilde{n}_s$

$$\chi \propto \int ds\, s^2 \frac{e^{-\Delta h s^{\tilde{\sigma}}}}{s^{\tau}} \propto \Delta h^{-1/2}, \tag{133}$$

and the order parameter density ψ (see Appendix A) is related to the first moment of $\tilde{n}_s$:

$$\psi \propto \int ds\, s \frac{e^{-\Delta h s^{\tilde{\sigma}}}}{s^{\tau}} \propto \Delta h^{1/2}. \tag{134}$$

Here we have kept G_s constant and used the fact that hyperscaling holds. This constraint is appropriate for the scaling events we are considering because these models self-organize to run at a fixed distance from the spinodal with $G_s \sim 3$–5 [3]. We can assume that the exponential in the integrals is approximately one until $s^{\tilde{\sigma}} \sim \Delta h^{-1}$. Hence,

$$\frac{3-\tau}{\tilde{\sigma}} = \frac{1}{2}, \tag{135}$$

and

$$\frac{\tau-2}{\tilde{\sigma}} = \frac{1}{2}. \tag{136}$$

Equations (135) and (136) yield $\tau = 5/2$ which apparently differs from the measured value of $x = 3/2$. However, the exponents obtained by this reasoning assume that the cluster distribution is obtained by tossing bonds between occupied sites. In the earthquake case the clusters are grown from a seed, the site that is brought to failure by a loader plate update. Because for a cluster of size s there are s places that could have been the seed, the number of such clusters is sn_s, where n_s is given in Eq. (130) [39]. Hence, $\tau = x+1$ and hence $x = 3/2$, in agreement with the simulations of models with long-range stress transfer. The scaling form for n_s with $x \approx 2$ for the usual Burridge-Knopoff model with short-range interactions is not understood.

There are other properties of the distribution of earthquakes in the Rundle-Jackson-Brown CA model that can be obtained from consideration of the clusters in Ising models near spinodals. We refer the interested reader to Ref. 3. The relation between the CA and Burridge-Knopoff models for long-range stress transfer is discussed in Refs. 66 and 67.

X. SUMMARY AND CONCLUSIONS

We have shown that the structure of the clusters near mean-field critical points and (pseudo)spinodals and its relation to thermal fluctuations is more complicated than the

corresponding relation in systems that are not mean-field and which obey hyperscaling. Moreover, these fundamental clusters and their structure have physical consequences which implies that the clusters are not only convenient mathematical constructs.

For (near-)mean-field systems the thermal fluctuations are generated by fluctuations in the number of fundamental clusters. Because the clusters in (near-)mean-field systems play a different role and appear to be real physical objects rather than just mathematical constructs, we refer to them as fundamental clusters. The probability of finding a fundamental cluster is not given by the Boltzmann factor because the cluster lifetime is much less than the decorrelation time. Their physical consequences are seen most clearly near the pseudospinodal in supercooled liquids where the relation between the measurement time and the lifetime of the fundamental clusters yields predictions for the behavior of the structure function that are confirmed by simulations on near-mean-field systems and are contrary to mean-field theory.

In addition to the applications of the cluster structure we have discussed, there are many other applications that have shed light on physical processes. These applications include:

1. The elucidation of the early time structure of systems undergoing spinodal decomposition and continuous ordering, including the understanding of why the linear theory of Cahn, Hilliard, and Cook [20, 21, 22] fails first at large momentum transfer [23, 24], the fractal structure of the mass distribution of early time spinodal decomposition [25], and a physical interpretation of the fermionic (Grassman) variables associated with a supersymmetric representation of the early stage continuous ordering [23, 24, 68].

2. The phase separation of polymer and solvent in the presence of gelation [69].

3. Possible precursors to nucleation near the pseudospinodal [12].

Future work includes the application of cluster methods to the study of precursors to large earthquakes in the CA and Burridge-Knopoff models, the study of heterogeneous nucleation near pseudospinodals, the investigation of fracture and the merging of microcracks, and the study of the crossover from the linear regime of spinodal decomposition and continuous ordering to nonlinear evolution.

APPENDIX A: PERCOLATION MAPPING

To obtain a deeper understanding of the structure of the fluctuations near the mean-field critical point, we map the Ising critical point (mean-field and non-mean-field) onto a percolation transition for a properly chosen percolation model [38]. We first describe the mapping introduced by Kasteleyn and Fortuin [70] of the s-state Potts model onto random bond percolation. The latter is defined on a lattice where all the sites or vertices are occupied with probability one, and the bonds are occupied with a probability p_b. Clusters are defined as a set of sites connected to each other by bonds and not connected to any other sites in the lattice [39].

The Hamiltonian for the s-state Potts model is

$$H_{\rm P} = -J_{\rm P}\sum_{i,j}(\delta_{\sigma_i\sigma_j} - 1) - h_{\rm P}(\delta_{\sigma_i 1} - 1), \tag{A1}$$

where σ_i specifies the state of site i, $J_{\rm P} > 0$ is the coupling constant, and $h_{\rm P}$ is the Potts field; the Kronecker delta $\delta_{\sigma_i\sigma_j} \neq 0$ only when sites i and j within the interaction range are in the same state. We first set $h_{\rm P} = 0$ and write the Boltzmann factor $e^{-\beta H_P}$ as

$$e^{-\beta H_{\rm P}} = \prod_{ij}\left[\delta_{\sigma_i\sigma_j} + e^{-\beta J_{\rm P}}(1 - \delta_{\sigma_i\sigma_j})\right], = \prod_{ij}\left[(1 - e^{-\beta J_{\rm P}})\delta_{\sigma_i\sigma_j} + e^{-\beta J_{\rm P}}\right]. \tag{A2}$$

We associate a bond with $\delta_{\sigma_i\sigma_j} = 1$ and the absence of a bond with $\delta_{\sigma_i\sigma_j} = 0$. With this association the generating function for the random bond percolation model is obtained by differentiating the free energy for the s-state Potts model with respect to s and then setting s equal to 1 [70]. There are many subtle mathematical points in the Kasteleyn-Fortuin proof of this relation, and we refer the reader to Ref. 70 and the references therein for the details. Because it will be needed to understand the structure of fluctuations, we demonstrate how the connection between the Potts model and percolation works.

The partition function $Z_{\rm P}$ is given by

$$Z_{\rm P} = \sum_{\sigma} e^{-\beta H_{\rm P}}. \tag{A3}$$

and the free energy in the canonical ensemble is $F_{\rm P}(\beta, s) = -k_B T \ln Z_{\rm P}$. If we differentiate $-\beta F_{\rm P}(\beta, s)$ with respect to s, we obtain

$$-\beta\frac{\partial F_{\rm P}(\beta, s)}{\partial s} = \sum_{\sigma}\frac{1}{Z_{\rm P}}\frac{\partial\, e^{-\beta H_{\rm P}}}{\partial s}. \tag{A4}$$

Setting $s = 1$ results in $H_{\rm P} = 0$ because there is only one Potts state; hence $Z_{\rm P} = s^N = 1$ for $s = 1$, where N is the number of sites in the lattice. Therefore

$$-\beta \frac{\partial F_{\rm P}(\beta, s)}{\partial s}\Big|_{s=1} = \frac{\partial}{\partial s} \sum_{\sigma} e^{-\beta H_{\rm P}}\Big|_{s=1}. \tag{A5}$$

The percolation generating function is the right-hand side of Eq. (A5). To understand this interpretation we consider several terms in Eq. (A5). We use Eq. (A2) for $e^{-\beta H_{\rm P}}$ and first consider the term $e^{-\beta J_{\rm P}}$ in each of the factors in the product; that is, we include no terms with $\delta_{\sigma_i \sigma_j}$. For a lattice with $c = qN/2$ total possible bonds, we find a contribution to $F_{\rm P}(\beta, s)$ of the form $s^N e^{-\beta J_{\rm P} cN}$. By differentiating with respect to s and setting $s = 1$, we obtain the contribution to the generating function $G_{\rm f}$:

$$G_{\rm f,1} = N e^{-\beta J_{\rm P} cN}. \tag{A6}$$

Because there are N sites and cN bonds, $G_{\rm f,\,1}$ can be interpreted as the mean number of single site clusters. That is, $e^{-\beta J_{\rm P} cN}$ is the probability that there are no bonds present.

We now consider a term from Eq. (A2) that includes only one delta function, which we take to be $\delta_{\sigma_1 \sigma_2}$. The contribution to $G_{\rm f}$ has the form

$$G_{\rm f,\,2}(p) = (1 - e^{-\beta J_{\rm P}}) s e^{-\beta J_{\rm P}(cN-1)} s^{N-2} = p_b (1 - p_b)^{cN-1} s^{N-1}, \tag{A7}$$

where we have associated the bond probability p_b with $1 - e^{-\beta J_{\rm P}}$. Differentiating with respect to s and setting $s = 1$ gives $N - 1$ for the number of clusters times the probability of such a configuration. There are $N - 2$ one-site clusters and one two-site cluster for a particular bond. The number of ways we can choose one bond is cN, so that the first two contributions to $\partial F(\beta, s)/\partial s$ are

$$G_{\rm f,\,1}(p) + G_{\rm f,\,2}(p) = N(1 - p_b)^{cN} + (N - 1) cN p_b (1 - p_b)^{cN-1}. \tag{A8}$$

If we continue in this manner, we would find that the terms we obtain are the number of clusters in a given configuration. The complete enumeration of the configurations will lead to an expression for the mean number of clusters as a function of p_b. Hence, the mean number of clusters can be written as

$$G_{\rm f}(p) = \sum_k <n_k>, \tag{A9}$$

where $<n_k>$ is the mean number of clusters with k sites.

To obtain the full generating function for random bond percolation, we must include the field $h_{\rm P}$ in the calculation of the free energy. We write

$$e^{-\beta H_{\rm P}} = \prod_{ij} \left[(1-e^{-\beta J_{\rm P}})\delta_{\sigma_i\sigma_j} + e^{-\beta J_{\rm P}}\right] \prod_{l} \left[(1-e^{-\beta h_{\rm P}})\delta_{\sigma_l 1} + e^{-\beta h_{\rm P}}\right]. \tag{A10}$$

The terms in the expansion of Eq. (A10) represent sets of connected sites (clusters) generated by $\delta_{\sigma_i\sigma_j}$. Terms of the form $\delta_{\sigma_k 1}$, where the index 1 labels one of the s possible states of a site, give no contribution to the derivative of the partition function with respect to s because the $\delta_{\sigma_k 1}$ term fixes all spins in a cluster to the Potts state labeled as 1. Hence there is no s dependence and no factor of s in the product, which equals s raised to the power of the number of clusters. From Eq. (A10) all clusters with a nonzero weight after differentiation with respect to s will have a field dependence of the form $e^{-nh_{\rm P}}$, where n is the number of sites in the cluster. If we resum as in Eq. (A8), we obtain [70]

$$G_{\rm f}(p_b, h_{\rm P}) = \sum_k <n_k> e^{-k\beta h_{\rm P}}. \tag{A11}$$

The reader might want to work out the generating function for small lattices to see how the sum in Eq. (A11) arises.

To investigate the mapping of the percolation model onto the Ising model, we consider the dilute s-state Potts [71] model with the Hamiltonian

$$\beta H_{\rm DP} = -\beta J_{\rm P} \sum_{ij} (\delta_{\sigma_i\sigma_j} - 1) n_i n_j - \beta h_{\rm P} \sum_i (\delta_{\sigma_i 1} - 1) n_i - K_{\rm LG} \sum_{ij} n_i n_j + \Delta \sum_i n_i. \tag{A12}$$

where $n_i = 1$ denotes that a particle (spin) occupies site i and $n_i = 0$ denotes an empty site. Hence there is a Potts interaction between occupied ($n_i = 1$) sites. The quantity

$$\beta H_{\rm LG} = -K_{\rm LG} \sum_{ij} n_i n_j + \Delta \sum_i n_i \tag{A13}$$

is the Hamiltonian for the lattice gas formulation of the Ising model [72], with $K_{\rm LG}$ the (dimensionless) coupling constant and Δ the chemical potential. In terms of the parameters in the Ising Hamiltonian $H_{\rm I}$,

$$\beta H_{\rm I} = -K_{\rm I} \sum_{ij} s_i s_j + \beta h_{\rm I} \sum_i s_i \tag{A14}$$

with $s_i = \pm 1$, we have [72]

$$K_{\rm LG} = 4K_{\rm I} \text{ and } \Delta = \beta h_{\rm I} + 2cK_{\rm I}. \tag{A15}$$

Differentiating the free energy constructed from $H_{\rm DP}$ with respect to s and setting $s = 1$ results in the generating function for correlated site random bond percolation for which occupied sites are distributed according to the lattice gas Hamiltonian in Eq. (A13) and bonds are thrown randomly with a probability p_b between pairs of occupied sites. To understand this connection we note that

$$Z_{\rm DP} = \sum_{\sigma_i \sigma_j n_i n_j} e^{-\beta H_{\rm DP}}, \tag{A16}$$

and

$$-\frac{\partial k_B T \ln Z_{\rm DP}}{\partial s}\Big|_{s=1} = \frac{\frac{\partial}{\partial s}(\sum_{\{\sigma_i\}\{n_i\}} e^{\sum_{ij} \beta J_{\rm P}(\delta_{\sigma_i\sigma_j}-1)n_i n_j + \beta h_{\rm P} \sum_i (\delta_{\sigma_i 1}-1)n_i} e^{-\beta H_{\rm DP}})\Big|_{s=1}}{Z_{\rm DP}}. \tag{A17}$$

If we write

$$\exp\left[\beta J_{\rm P}(\delta_{\sigma_i\sigma_j}-1)n_i n_j\right] = [(1-e^{-\beta J_{\rm P}})\delta_{\sigma_i\sigma_j} + e^{-\beta J_{\rm P}}]n_i n_j + (1 - n_i n_j), \tag{A18a}$$

and

$$\exp\left[\beta h_{\rm P} \sum_i (\delta_{\sigma_i 1} - 1)n_i\right] = (1-e^{-\beta h_{\rm P}})\delta_{\sigma_i 1} n_i + e^{-\beta h_{\rm P}} n_i + (1 - n_i), \tag{A18b}$$

we can use the same arguments that we gave for random percolation to show that the expression in Eq. (A17) leads to the generating function for correlated site random bond percolation where the occupied sites are distributed according to the lattice gas Boltzmann factor constructed from the Hamiltonian in Eq. (A13) [71]. Note that for this model the sum over the s states of the Potts spin is one for all s if site i is empty.

We now consider the Hamiltonian $H_{\rm DP}$ in Eq. (A12). We set $h_{\rm P} = h_I = 0$, and hence $\Delta = 2qK_{\rm I} = qK_{\rm LG}/2$ from Eq. (A15) and let $J_{\rm P} = K_{\rm LG}/2$ in Eq. (A12). Then $H_{\rm DP}$ becomes

$$\beta H_{\rm DP} = -\frac{K_{\rm LG}}{2}\sum_{ij}(\delta_{\sigma_i\sigma_j}-1)n_i n_j - K_{\rm LG}\sum_{ij} n_i n_j + \frac{K_{\rm LG}}{2}\sum_{ij}(n_i + n_j). \tag{A19}$$

Suppose that for a pair of sites within the interaction range, either both sites are empty or both sites are filled, but the spins are in the same Potts state. When both sites are empty, there is only one Potts configuration. When both sites are occupied and in the same Potts state, there are s configurations, Hence, there are $s + 1$ configurations with $H_{\rm DP} = 0$. Now

consider the case where either one site is occupied and one empty or both sites are occupied, but the spins are in different Potts states. In this case the contribution of this pair to $H_{\rm DP}$ is $K_{\rm LG}/2$, and the number of ways this combination can be obtained is $(s+1)s$. These considerations imply that the dilute s-state Potts model at $J_{\rm P} = K_{\rm LG}/2$ is equivalent to a pure $(s+1)$-state Potts model with the Hamiltonian

$$\beta H_{{\rm P},\,(s+1)} = -\frac{K_{\rm LG}}{2}(\delta_{\sigma_i\sigma_j} - 1), \tag{A20}$$

where σ_i can be in $s+1$ states. The $s = 2$ Potts model is the lattice gas model in Eq. (A12) with $\Delta = cK_{\rm LG}/2$. That is, for $\beta J_{\rm P} = K_{\rm LG}/2 = 2K_{\rm I}$ and $h_I = h_{\rm P} = 0$, the Hamiltonian of the dilute s-state Potts model is the same as the Hamiltonian of the $(s+1)$-state pure Potts model. In the limit $s \to 1$, the $(s+1)$-state pure Potts model is the lattice gas model.

In this formulation we can write the Boltzmann factor as

$$e^{-\beta H_{{\rm P},\,(s+1)}} = \prod_{ij}[(1 - e^{-K_{\rm LG}/2})\delta_{\sigma_i\sigma_j} + e^{-K_{\rm LG}/2}] \tag{A21}$$

$$= e^{-K_{\rm LG}cN/2}\prod_{ij}\Big[\frac{1 - e^{-K_{\rm LG}/2}}{e^{-K_{\rm LG}/2}}\delta_{\sigma_i\sigma_j} + 1\Big]. \tag{A22}$$

Clearly the singular behavior of the free energy comes from the terms in Eq. (A22) contained in the product over lattice sites. This product has the form of $\prod_{ij}[f_{ij}+1]$, where we associate a graph or cluster with a product of the f_{ij} summed over Potts states. These clusters are the same as the percolation clusters because the sites are connected by $\delta_{\sigma_i\sigma_j}$ bonds. The linked cluster theorem [73, 74] states that the singular part of the free energy $F_{\rm P,\,sing}$ is the sum over all *connected* graphs in the thermodynamic limit. Connected graphs are those in which all points or vertices of the graph are connected by an f_{ij} bond. Because the graphs are connected, the sum over Potts states results in a factor of s, independent of the size or structure of the graph. Therefore the derivative of $F_{\rm P,\,sing}$ with respect to s results in the same sum without the overall factor of s. Thus

$$2\frac{dF_{\rm P,\,sing}}{ds}\Big|_{s=1} = F_{\rm P,\,sing}\Big|_{s=1}, \tag{A23}$$

which implies that the percolation transition and the Ising critical point occur at the same temperature and have the same critical exponents. The amplitudes of the singular quantities differ by a factor of two. If instead of the Hamiltonian in Eq. (A12), which defines clusters

as consisting of only occupied sites, we add a term to the Hamiltonian of the form

$$\begin{aligned}\beta H_{\mathrm{DP,empty}} &= -\beta J_{\mathrm{P}} \sum_{ij} (\delta_{\sigma_i \sigma_j} - 1)(1 - n_i)(1 - n_j) \\ &\quad - K_{\mathrm{LG}} \sum_{ij} (1 - n_i)(1 - n_j) + \Delta' \sum_{i} (1 - n_i), \end{aligned} \tag{A24}$$

we can define clusters between "empty" sites. In this way the singular part of the free energy will be identical to the mean number of clusters for $s \to 1$ and $h_{\mathrm{P}} = 0$. The same result was obtained using renormalization group techniques in Ref. 38. The mapping of the Ising model onto a properly chosen percolation model is not restricted to nearest neighbor interactions. If the Ising interaction has a range R, we need only to choose the Potts interaction to also have a range R, which means that the bond between sites is randomly placed between any two spins within the interaction range.

We now consider the mapping of a thermal problem onto a percolation model near the spinodal. This mapping will require a slightly different approach. We again begin with the dilute s-state Potts model. The Hamiltonian is the sum of H_{DP} and H_{LG} from Eqs. (A12) and (A13). This Hamiltonian can be put into a continuum form using the Gaussian transformation [31, 75]. Because we are interested in the mean-field limit, the Landau-Ginzburg-Wilson Hamiltonian is the free energy. We have from Refs. 25 and 75

$$\begin{aligned} F_{\mathrm{DP}}(\zeta, \phi) = & \int d\mathbf{x} \Big[\frac{1}{2} s(s-1) \big(R \nabla \zeta(\mathbf{x}) \big)^2 - r_1 s(s-1) \zeta^2(\mathbf{x}) - h_{\mathrm{P}}(s-1)\zeta(\mathbf{x}) \\ & + \frac{w_1}{4!} s(s-1)(s-2)(s-3)\zeta^3(\mathbf{x}) + \frac{w_2}{2} s(s-1)\zeta^2(\mathbf{x})\phi(\mathbf{x}) \Big] + F(\phi), \end{aligned} \tag{A25}$$

where $F(\phi)$ is the free energy in Eq. (48). The constants r_1, w_1, w_2, and ϵ in Eq. (48) can be written as functions of J, K_{LG}, and c. The global percolation order parameter ζ is the probability that a spin in the stable phase direction belongs to the infinite cluster of occupied sites. As for a discrete system, the percolation model is obtained by differentiating F_{DP} with respect to s and setting $s = 1$ [75].

To map the thermal problem near the spinodal onto the percolation problem, we rewrite Eq. (48) as in Eq. (52) with $\Delta h = h_s - h_I$. We equate the functional derivative of $dF_{\mathrm{DP}}(\zeta, \phi)/ds|_{s=1} = F_{\mathrm{P}}$ with respect to $\zeta(\mathbf{x})$ to the functional derivative of $F(\psi)$ with respect to ψ. That is,

$$\frac{\delta F_{\mathrm{P}}}{\delta \zeta} = -R^2 \nabla^2 \zeta(\mathbf{x}) - 2 r_1 \zeta(\mathbf{x}) + \frac{w_1}{3} \zeta^2(\mathbf{x}) + w_2 \zeta(\mathbf{x}) \phi(\mathbf{x}) - h_{\mathrm{P}}, \tag{A26}$$

and

$$\frac{\delta F(\psi)}{\delta \psi} = -R^2 \nabla^2 \psi(\mathbf{x}) + 2\lambda_1 \Delta h^{1/2} \psi(\mathbf{x}) - 3\lambda_2 \psi^2(\mathbf{x}) + 4\lambda_3 \psi^3(\mathbf{x}) \tag{A27}$$

must be equal. This condition implies that $-2(r_1 - \frac{w_2}{2}\phi_s)\zeta + \frac{w_1}{3}\zeta^2 - h_{\rm P}$ must be the same as $2\lambda_1 \Delta h^{1/2}\psi - 3\lambda_2 \psi^2$. We dropped the ψ^3 term in the Euler-Lagrange equation because $\psi \sim \Delta h^{1/2} \ll 1$ as we shown in Secs. VI and XI.

At the spinodal ($\Delta h = 0$) we identify ζ with $-\psi$ and require that $h_{\rm P} = 0$, $w_1/3 = -3\lambda_2$, and $r_1 = w_2\phi(\mathbf{x})/2$. With these equalities the solutions of Eqs. (A26) and (A27) with $\delta F_P/\delta\zeta(\mathbf{x}) = \delta F(\psi)/\delta\psi(\mathbf{x}) = 0$ are identical. If we write the parameters in Eqs. (A26) and (A27) in terms of the parameters $J_{\rm P}$, $K_{\rm LG}$, and c of $H_{\rm DP}$ in Eqs. (A12) and (A13), we obtain

$$\beta J_{\rm P} = 2K_I(1-\rho), \tag{A28}$$

where the density $\rho = (1+m)/2$ and ϕ is proportional to m, the magnetization per spin. Because we are interested in the coincidence of the spinodal with a percolation transition, we can set $\phi(\mathbf{x}) = \phi_s$, the value of the order parameter at the spinodal.

To define the bond probability in terms of a physically meaningful quantity, we need to obtain the proportionality factor between ϕ_s and m_s, the value of the magnetization at the spinodal. To do so we use the relation between the Ising model and the ϕ^4 theory generated by the Hubbard-Stratonovich transformation [31]. In particular, the value of ϕ_s is [75]

$$\phi_s = \pm \frac{[(cK_I - 1)c]^{1/2}}{c^2 K_I}. \tag{A29}$$

When $T \to 0$ or equivalently $K_I \to \infty$, $m_s \to 1$ as can be seen by noting that the magnetic field is divided by T in the Boltzmann factor. Because $\phi_s \to 1/cK_I^{1/2}$ as $K_I \to \infty$, we have

$$m_s = cK_I^{1/2}\phi_s, \tag{A30}$$

so that $m_s \to 1$ as $K_I \to \infty$. If we use Eq. (A30), we obtain the expression for the bond probability that maps the percolation model onto the spinodal

$$p_b = 1 - e^{-\beta J_{\rm P}}, \tag{A31}$$

where $J_{\rm P}$ is given in Eq. (A28). The validity of this mapping was demonstrated numerically in Refs. 10, 12, and 76.

The interpretation of this mapping is that the spinodal curve is the locus of a set of percolation transitions. If the spinodal curve is approached with $h \neq 0$, there is a transition

to a spanning cluster in the stable phase direction at the spinodal. We stress that this result is correct only in the mean-field limit, $G_s \to \infty$.

The various mappings we have discussed imply that the free energy of the lattice gas model is isomorphic to the generating function for correlated site random bond percolation. For $h_{\rm P} = h_I = 0$, the generating function is the mean number of clusters.

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